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学位論文内容の要旨

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学位論文題名

A study on the particulate transport in the sea with cosmic ray produced Be-7 and other natural radionuclides.

(宇宙線生成核種の ^7Be および他の天然放射性核種による海洋での物質の粒状物による輸送に関する一研究)

近年の人類活動は、産業廃棄物、下水、煤煙などを通して多くの化学物質を環境に放出している。これら人類起源物質の流入にともなう物質循環の変化は海洋の化学組成に大きな影響を与える可能性があるはかりでなく、そこに棲む多くの生物にも重大な影響を与える場合がある。特に重金属は水俣病に代表されるように人類にまでその影響が及んでいるが、それがごく微量で海の生態系に大きな影響を与えることと考えられる。今後、ますます食料資源を海に求めるようになるであろうし、またマンガン団塊などの海の鉱物資源の利用が始まるであろう。海洋での重金属の挙動を正確に把握しておくことはこれらの諸問題を解決し、我々人類の将来を設計するために不可欠のことと思われる。

本研究では、宇宙線で生成する ^7Be 、天然の放射性核種であるTh同位体、 ^{210}Po 、 ^{210}Pb をとりあげた。これらはいずれも金属の放射性同位体であるため、これらを用いて鉄、マンガン、アルミニウムその他重金属の環境における挙動を解明することができると考えられる。放射性核種を利用する利点はまず第一に微量で正確に

測定できることである。通常の安定な重金属では試料採取から測定までの間に汚染の除去が大問題となっており、特に温度、圧力に影響されず一定の速さで親核種から生まれ、一定の速さで放射壊変して減少してゆくために、それらの存在量から直ちに供給、除去の速さを見積ることができると考えられる。通常の観測では時間変化を把握しきれない変化速度を見積ることはできない。

重金属の海洋での分布が生物の活動と密接に関係していることは多くの研究者によってすでに指摘されている。しかしながら、この結論は海水中に溶けている物質についての結果から得られたものであり、例えば海洋における沈降粒子束を測定するなど、それを直接的に解明したものではない。

本研究では海水について研究することのほか、セジメント・トラップを一定期間海洋中に設置し、実際に物質を下方に運搬している粒子を捕集し、その化学的あるいは放射化学的に分析して、各成分の挙動を明らかにしようと試みた。実験海域としては汚染されていない内湾として噴火湾、汚染されている内湾として東京湾、亜外洋域の相模湾と尻屋崎60マイル沖、外洋域として北部北太平洋を選んだ。ここで得られたセジメント・トラップ試料と海水試料中の ^7Be 、 ^{234}Th 、 ^{210}Po 、 ^{210}Pb 、 ^{228}Th 、 ^{230}Th 、 ^{232}Th の測定を行った。得られた結果は以下の通りである。

1. 噴火湾における ^7Be で得られたBeの平均滞留時間は90日～160日で、これは ^{234}Th で得られた値とほぼ一致した。すなわち、これらは

同様な運搬機構によって、海洋から比較的速やかに除かれていることを示している。

2. 海水中の ^{90}Be の鉛直分布は粒子による鉛直輸送の影響を強くうけている。従って ^{90}Be を用いて表面水中の鉛直拡散係数を算出するためには粒子による沈降の項を入れなければならぬ。

3. セジメント・トラップで観測された粒子束は海水中濃度を解析して得られた正味の除去量より大きく、粒子は単に一方的に下方に沈降するものではないことを示している。

4. セジメント・トラップ実験の結果より海水中の重金属は容易に大粒子になったり小粒子になったりする粒子によって運搬されている可能性があることがわかった。

5. 小粒子化した粒子の沈降速度は無視できるほど小さいので、多くの重金属は溶解したくても粒子態のままて海水中を上昇しているものと考える。その程度は海域によって大きく異なるが、外洋ではむしろ海底に近づくほど大きくなる。

現在、多くの研究者は海洋の重金属成分はフローラ・フタニの糞粒のような大きな粒子によって不可逆的過程によって海洋からとりのぞかれていると考えているが、このような簡単なメカニズムではこれら

金属成分の挙動を説明することはできない。同じ大粒子でもマリン・スローの方が大きく成長したり小さくこわれたりする子ので重金属の運搬役として考えやすいこともわかっている。

A STUDY ON THE PARTICULATE TRANSPORT IN THE SEA WITH COSMIC RAY
PRODUCED BE-7 AND OTHER NATURAL RADIONUCLIDES

BY

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ABSTRACT

Almost all the parts of chemical substances introduced into the oceans are removed by particulate transport downward to the bottom. However, our knowledge on the conveyers of these substances, such as the chemical composition of sinking particles, their sinking rate, the mechanism of their formation, etc. is quite limited. Among such substances, heavy metals are of special interest because of their high toxicity. Recently, these metals are supplied with an increasing rate to the oceans from the burning of fossil fuels, the industrial waste and others.

So as to study the behavior of heavy metals in the oceans, sinking particles were directly collected with sediment traps in coastal sea (Funaka Bay, Tokyo Bay), hemipelagic sea (Sagami Bay) and pelagic sea (the North Pacific), and Be-7, Th-234, Po-210, Pb-210 and long lived Th isotopes in the settling particles were determined in order to use them as timers (geochronometer) of the settling.

The results obtained are as follows.

1. The residence time of Be determined with Be-7 in Funaka Bay is 90 - 160 days, which is approximately equal to that of Th-234.
2. The vertical distribution of Be-7 in the surface ocean is strongly controlled by particulate transport. The eddy diffusion coefficient of surface water estimated without taking particulate transport of the nuclide into consideration in the previous studies may not be real.

3. The observed flux with sediment traps does not give the net removal flux of chemical substances in seawater .
4. The heavy metals in seawater are mainly transported downward by the particles which easily grow up to large aggregates with significant velocities and break down to small particles of colloidal sizes while sinking.
5. The small colloidal particles move up together with undulation of water in the ocean. Thus, the "upward flux" of particulate matter exists probably everywhere in the oceans.

At present, many investigators believe that fecal pellets mainly play an important role on the transport of heavy metals from the surface to the deep in the oceans. However, the results obtained in this research show that large aggregates formed from suspended matter in sea-water are more important for the vertical transport of these metals in the oceans. Marine snow seems to be responsible for the transport.

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CHAPTER I

GENERAL INTRODUCTIONI - 1 Introduction

From the last century, the population pressure on the earth is increasing from day to day. The time is going to come in the nearer future when almost food and industrial resources are to depend on marine products. The assessment of man's impacts such as fishing, collection of seaweed, mining, and pollution on the chemistry and biology in the oceans can be a part of the important studies in fisheries science. Among man's introduced substances into the oceans, heavy metals, above all, must be studied extensively on their chemical nature in the oceans and their high toxicity to the marine ecosystem.

Seawater is not merely saline water in a bottle, since many kinds of organisms are living and enormous chemical reactions are taking place. Huge amount of substances is supplied continuously to the oceans and many compounds are produced there. The products are taken off from seawater (precipitation, rock-water interaction, gravitational settling, etc.). As a result of a balance among these processes, a certain composition of seawater is maintained. This is the reason why the oceans are often likened to a chemical plant.

The residence times of heavy metals contained in seawater such as iron, cobalt, manganese, lead and thorium are much shorter than those of major elements such as sodium, calcium and magnesium.

Judging from the residence times of these heavy metals in the oceans, we can easily obtain their sinking rates to be a few meters per day. The settling of these metals alone can be hardly explained their high sinking rates expected above. Almost all investigators have come to a conclusion that particulate materials in seawater contribute to the vertical transport of such heavy metals in the oceans. However, the following questions are still remained, which we must study to find their answers.

1. What kind of particles are responsible for the removal of heavy metals from seawater?
2. How fast do they sink to the ocean floor?
3. What is their chemical composition?

I - 2 Naturally occurring radioisotopes

The presently known natural radioactive nuclides are listed in Table I - 1. U-238, U-235 and Th-232 are the precursors of successive decays. Namely, we call them as the uranium series, the actinium series and the thorium series, respectively. The decay schemes for each decay series are shown in Figure I - 1, I - 2 and I - 3.

Many useful nuclides for the study of geochemical processes belong to those decay series. For example, daughters of Rn-222 are used for the study of the atmospheric aerosol. Th-230 and Pa-231 are used as a geochronometer of the deep sea sediments and Ra-228 and Ra-226 are used as a monitor for horizontal and vertical mixing of sea-water. The some results of the research of particulate removal process in the oceans studied with these elements will be shown in the following sections.

Among the nuclides which are not included in the decay series, K-40, Rb-87 and Sm-147 are sometimes used for the geochronology of rock and meteorite. The remainders are not used as geochemical monitors at present.

I - 3 Study of natural radioisotopes as tracers of heavy metals in sea water.

Th isotopes. Po-210 and Pb-210 have been given an attention to study removal mechanisms in the oceans, since these elements are expected to have a similar chemical nature.

The studies of Th in sea water (Bhat et al., 1969; Matsumoto, 1974; Broecker et al., 1973; Minagawa and Tsunogai, 1980) have revealed that Th in the surface water has a shorter residence time than that of the usual surface water and the residence time of Th in the shallow water is shorter than that in open sea. The same results have been observed for Pb-210 and Po-210. Aller and Cochran (1976) have obtained the shortest residence time of Th-234 of only one day in Long Island Sound, U.S.A.

So as to explain high removal rates for these elements, Broecker et al. (1973) demonstrated that the removal rate of Th-228 strongly depends on biological activity, especially uptake by plants in their quest for phosphate. Nozaki and Tsunogai (1973), Benninger (1976), Kharkar (1976) and Turekian (1977) pointed out that the uptake of these elements by living phytoplankton alone is not significant for the removal of Th-234, Th-228, Pb-210 and other elements.

Spencer et al. (1978) and Brewer et al. (1980) asserted that the removal of such elements by rapidly sinking particles such as zooplankton fecal pellets is more important than that of aggregates of suspended matter. Beasley et al. (1976) also pointed out that fecal pellets are one of the important conveyers for the vertical transport of Po-210 and Pb-210 in sea water. Tsunogai and Minagawa (1978) have demonstrated the downward transport by the sinking of large aggregates, "Marine snow" which can grow up from particles of colloidal sizes containing metals such as Th, Pb and Fe, and sometimes break down to small particles while sinking down through seawater.

I - 4 Cosmic ray produced isotopes.

Nuclear transformations by cosmic ray radiation take place in the atmosphere, since most of the cosmic rays is dissipated there, especially in the stratosphere. Figure I - 4 shows the altitude dependence of the vertical flux of particles, which consist the cosmic rays, at 45° of latitude reported by Peters (1958). Almost all the transformations are produced by nucleons. The contribution of mesons and their decay products, i.e. muons, electrons and gamma rays are relatively small. Multiplying the nuclear interaction cross-section for each kind of particles with each flux at the point where the flux reaches its maximum value, Lal and Peters (1967) estimated the contribution of protons, γ -rays and muon to the nuclear disintegrations:

$$\text{protons} : \gamma\text{-rays} : \text{muons} = 100 : 5 : 0.025$$

The atmospheric target nuclei are N, O, Ar and Kr. Since the Ar and Kr contents in the atmosphere are low, 1 % or less by volume, isotopes of atomic weight less than 16 are produced from interactions with N and O nuclei. Major cosmic ray produced nuclides which have half-lives longer than 1 day are listed in Table I - 2.

The fallout patterns for these nuclides show a strong latitudinal dependency, since the cosmic ray intensity and the altitude of the tropopause vary with latitude. Additionally, the discontinuity of the tropopause in the temperate zone lead to the transfer of air between ^{the}stratosphere and ^{the}troposphere and seems to show a seasonal variation.

Many of the nuclides become oxides soon after creation, except noble gases He, Ar and Kr. Among oxides, only CO₂ is a gas and remains free, while the others attach to aerosols in the atmosphere in a very short time. The size distribution of aerosols in the atmosphere and the attachment of radioisotopes to them have been studied extensively by Junge (1963). He suggested that the attachment of these nuclides to aerosols occurs mainly to particles of less than one micron diameter, whose gravitational settling is not important. Therefore, cosmic ray produced isotopes attached to aerosols behave together with air. Removal of these nuclides from the atmosphere occurs with the atmospheric precipitations which collect aerosols during rainout and washout processes. On the other hand, the cosmic ray produced gases such as radiocarbon are removed by much slower processes, namely, molecular exchange at the surface of the oceans and the uptake by plants during photosynthesis on land.

Thus, cosmic ray produced nuclides could be classified into two groups with the differences in their sink functions.

I - 5 History of the study of Be-7 and its applications for the tracer studies in the oceans.

The cosmic ray produced radioisotope, Be-7 (half-life; 53 days), may be useful to trace various geochemical processes occurring in the hydrosphere with a time scale from a few tens to about 200 days. Lal et al. (1963) calculated the mean concentration of Be-7 in the surface sea water (75 m) from its production rate in the atmosphere to be 180 dpm/1000l and reported 10 to 70 dpm/1000l in the surface water of the Indian Ocean. They collected Be-7 in the surface water on sponge or jute impregnated with ferric hydroxide. Silker et al. (1973) and Young and Silker (1974) energetically carried out the measurement of Be-7 in the surface sea water for the estimation of the deposition rate of Be-7 from the atmosphere and the vertical eddy diffusion coefficient of surface sea water.

Merril et al. (1960) estimated the residence time of Be in the oceans to be 150 years. This value is very close to that of quickly precipitating elements such as Al, Fe, Ti and Th as discussed by Tsunogai and Minagawa (1978). This fact suggests that Be-7 relates to the particulate removal on the oceans. If it is the case, Be-7 can be used as a good tracer to study the vertical transport process by particulate matter in the surface layer of the oceans. The strongest point of Be-7 as the tracer is that its source is confined to the surface. Therefore, the transport rates of heavy metals from the surface to the deeper

layer might be estimated directly from its vertical profile. Bishop et al. (1977) collected a large amount of particulate matter in the sub-surface water by a Large Volume in situ Filtration System (LVFS) and suggested the particulate removal of Be-7 is due to the surface adsorption to the small suspended matter rather than the active biological uptake, since the fraction larger than 58 microns exhibited no measurable Be-7 activity.

I - 6 Objectives of this dissertation.

As mentioned in the previous sections, the amounts of heavy metals introduced into the oceans by human activities are increasing and the consequent influence introduced in the marine ecosystem is serious because of their high toxicity. In order to make clear the behaviours of heavy metals in the oceans, naturally occurring radioisotopes which have similar chemical nature to them have been utilized, since these nuclides can be used as timers of naturally occurring phenomena (geochronometer) and this is the strongest point of their use. Additionally, the nuclides once caught in particulate matter are not only to be tracers of such metals but also to be tracers of all the chemicals in the particles.

Many observations have shown that the removal rates of these elements strongly depend on biological activity in the oceans. However, the particles which actually remove reactive chemical compounds have not been directly studied. I used sediment traps to collect the settling particles transporting such compounds to the deep layer and measured radioisotopes

having respective source functions and half-lives in seawater. The radioisotopes measured in this research are listed in Table I - 3. From the results in this research, I will conclude that the large aggregates formed from small suspended particles play an important role for the vertical transport of insoluble heavy metals in the sea and that it is necessary that the large aggregates should break down to small sizes and reform actively within a very short period while sinking. This means that the suspended particles which have a negligible slow sinking velocity are also closely related to the active removal of such heavy metals from seawater. If "Marine snow" repeats actively to transform its particle sizes between the large aggregates which can settle downward and the small colloidal particles which can not sink, I will propose a suggestion that "Marine snow" should be such a conveyer of heavy metals in the oceans.

U 92	U^{238}, U_I (uranium I) 4.49×10^9 years		U^{234}, U_{II} (uranium II) 2.48×10^5 years				
Pa 91	α	Pa^{234}, UX_2 1.18 minutes β	Pa^{234}, UZ 6.7 hours β	α			
Th 90	Th^{234}, UX_1 (uranium X ₁) 24.1 days		Th^{230}, Io (ionium) 8.0×10^4 years				
Ac 89			α				
Ra 88			Ra^{226}, Ra (radium) 1622 years				
Fr 87			α				
Rn 86			Rn^{222}, Rn (radon) 3.825 days				
At 85			α	At^{218} 2 seconds			
Po 84			Po^{218}, RaA (radium A) 3.05 minutes α	Po^{214}, RaC' (radium C') 1.6×10^{-4} second		Po^{210}, RaF (polonium) 138.4 days	
Bi 83			α (99.98%) Bi^{214}, RaC (radium C) 19.7 minutes β	Bi^{214}, RaC (99.96%) α	Bi^{210}, RaE (radium E) 5.0 days β		
Pb 82			Pb^{214}, RaB (radium B) 26.8 minutes α (0.04%) Pb^{210}, RaD (radium D) 22 years β		α ($5 \times 10^{-5}\%$) Pb^{206}, RaG (stable lead isotope)		
Tl 81			Tl^{210}, RaC'' (radium C'') 1.32 minutes β		Tl^{206}, RaE'' (radium E'') 4.19 minutes β		

Figure I -1 The uranium series
(from Friedlander and Kennedy, 1955)

90	Th	Th ²³² , Th (thorium) 1.39 × 10 ¹⁰ years		Th ²²⁸ , RdTh (radiothorium) 1.90 years		
89	Ac		Ac ²²⁸ , MaTh ₂ (mesothorium 2) 6.13 hours			
88	Ra	Ra ²²⁸ , MaTh ₁ (mesothorium 1) 6.7 years		Ra ²²⁴ , ThX (thorium X) 3.64 days		
87	Fr					
86	Rn			Rn ²²⁰ , Th (thoron) 54.5 seconds		
85	At				At ²¹⁶ 3 × 10 ⁻⁴ second	
84	Po			Po ²¹⁶ , ThA (thorium A) 0.158 second		Po ²¹² , ThC' (thorium C') 3.0 × 10 ⁻⁷ second
83	Bi				Bi ²¹² , ThC (thorium C) 60.5 minutes	
82	Pb			Pb ²¹² , ThB (thorium B) 10.6 hours		Pb ²⁰⁸ , ThD (stable lead isotope)
81	Tl				Tl ²⁰⁸ , ThC" (thorium C") 3.1 minutes	

Figure I - 2 The thorium series

(from Friedlander and Kennedy, 1955)

92	U	U ²³⁸ , AcU (actinouranium) 7.13 × 10 ⁸ years				
91	Pa		Pa ²³¹ , Pa (protactinium) 3.43 × 10 ⁴ years			
90	Th	Th ²³¹ , UY (uranium Y) 25.6 hours		Th ²²⁷ , RdAc (radioactinium) 18.6 days		
89	Ac		Ac ²²⁷ , Ac (actinium) 22.0 years			
88	Ra			Ra ²²³ , AcX (actinium X) 11.1 days		
87	Fr		Fr ²²³ , AcK (actinium K) 21 minutes			
86	Rh			Rn ²¹⁹ , An (actinon) 3.92 seconds		
85	At		At ²¹⁹ 0.9 minute		At ²¹⁵ 10 ⁻⁴ second	
84	Po		Po ²¹⁸ , AcA (actinium A) 1.83 × 10 ⁻³ second		Po ²¹¹ , AcC' (actinium C') 0.52 second	
83	Bi		Bi ²¹⁸ 8 minutes		Bi ²¹¹ , AcC (actinium C) 2.16 minutes	
82	Pb		Pb ²¹¹ , AcB (actinium B) 36.1 minutes		Pb ²⁰⁷ , AcD (stable lead isotope)	
81	Tl			Tl ²⁰⁷ , AcC" (actinium C") 4.79 minutes		

Figure I - 3 The actinium series

(from Friedlander and Kennedy, 1955)

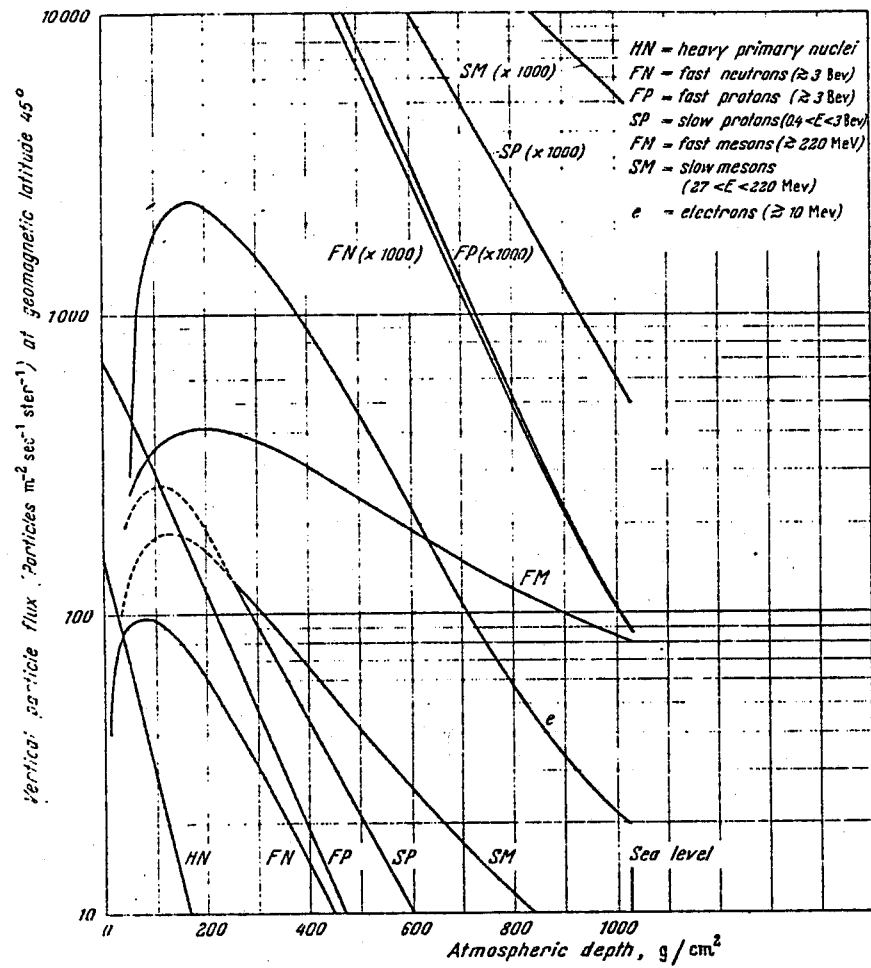


Figure I - 4.

Composition of cosmic radiation as a function of atmospheric depth. The flux values refer to geomagnetic latitude 45° and longitude 80° west. (from Peters, 1958)

Table I - 1. Naturally occurring radioisotopes (excluding cosmic ray produced ones). (from Friedlander and Niller, 1964)

Nuclide	Type of disintegration	Half-life (years)	Isotopic abundance (%)
U-238*	α	4.51×10^9	99.27
U-235*	α	7.13×10^8	0.72
Th-232*	α	1.39×10^{10}	100
K-40	β, EC	1.27×10^9	0.012
V-50	β, EC	6.0×10^{15}	0.24
Rb-87	β	5.7×10^{10}	27.9
In-115	β	5×10^{14}	95.7
Te-123	EC	1.2×10^{13}	0.87
La-138	EC, β	1.1×10^{11}	0.089
Ce-142	α	5×10^{15}	11.07
Nd-144	α	2.4×10^{15}	23.85
Sm-147	α	1.1×10^{11}	14.97
Gd-152	α	1.1×10^{14}	0.20
Lu-176	β	3×10^{10}	2.59
Hf-174	α	2×10^{15}	0.18
Re-187	β	6×10^{10}	62.9
Pt-190	α	7×10^{11}	0.013

* Precursors of successive decay chains

Table 1-2 Major cosmic ray produced isotopes in the atmosphere.
(Lal and Peters, 1957)

Isotope	Half-life	Main radiation	Main target nuclide(s)
Be-10	2.5×10^6 y	β^-	N, O
Al-26	7.4×10^5 y	β^+	Ar
Cl-36	3.1×10^5 y	β^-	Ar
Kr-81	2.1×10^5 y	K-X ray	Kr
C-14	5730 y	β^-	N, O
Si-32	710 y	β^-	Ar
Ar-39	270 y	β^-	Ar
H-3	12.3 y	β^-	N, O
Na-22	2.6 y	β^+	Ar
S-35	87 d	β^- , γ	Ar
Be-7	53 d	γ	N, O
Ar-37	35 d	K-X ray	Ar
P-33	25 d	β^-	Ar
P-32	14.3 d	β^-	Ar

Isotopes which have a half-time longer than 1 day are listed in this table.

Table I - 3. Rough classification of radioisotopes
used in this research according to their
sources

Sites of origin of radioisotopes	Nuclides
Atmosphere	Be-7, Po-210*, Pb-210*
Sea water	Th-234, Po-210, Pb-210, Th-230, Th-228
Land	Th-232

*Pb-210 and Po-210 in the atmosphere are produced by the decay of Rn-222 emanated from the earth's surface. A part of these nuclides is also produced from Ra-226 dissolved in sea water.

CHAPTER II

Vertical transport mechanisms of heavy metals in coastal sea.

The hydrography of Funka Bay has been established well by Ohtani and Kido (1980). The cold water of the Oyashio flows into this Bay in early spring. In summer, the water in the Bay is 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 winter. This water mass stays in the Bay till the water of the Oyashio flows in. I determined some radioisotopes in sea water and in the settling material collected with sediment traps. Figure II - 1 shows the locations of observed stations in Funka Bay. Sediment trap experiments were carried out at St. P. For comparison, the sampling of seawater and the collection of the settling matter with sediment traps were carried out also in Tokyo Bay.

II - 1 Analytical procedure

Th-234, Po-210 and Pb-210 in sea water were co-precipitated with ferric hydroxide and calcium carbonate from 50-liters of seawater after the solution acidified with nitric acid containing Th-232, Po-208 and stable Pb was kept as it stood for about 6 hours. After depositing Po 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 Po-210, a daughter of Pb-210. The Pb-210 activity was calculated from the Po-210 activity grown during storage. Th was electro-

deposited 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 directly from the sea was poured into a 500-liter polyethylene container on the vessel and analyzed for Be-7. Be-7 in the sampled seawater acidified with 1.5 l of conc. H_2SO_4 containing stable Be of 25 mg was co-precipitated with ferric hydroxide and calcium carbonate. The precipitates separated with decantation were dissolved with 3 l of 7.5 N HNO_3 and the obtained solution was boiled till dissolved carbon dioxide was completely evaporated off. 500 ml of conc. ammonia water was then added to the solution and the formed precipitate of hydroxide was filtered. The precipitate was dissolved with 200 ml of 6N HCl and the solution was shaken with 100 ml of methyl isobutyl ketone to extract almost all Fe. The separated water phase was completely evaporated to dryness on a hot plate and 10 ml of 0.5N HCl and 200 ml of 20 % NaOH solution were added. Although Be was left in the solution, 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 was adjusted pH to 9.6 with ammonia water to precipitate beryllium hydroxide. The precipitate collected on a filter was dissolved with 50 ml of 0.5 N HCl. 1 g of EDTA-2Na salt was added and the solution was adjusted pH to 5.3. After addition of 10 ml of acetate buffer solution (pH 5.3),

25 ml of acetyl acetone and 100 ml of benzene, the solution

was shaken for ten 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 the determination of chemical yield of Be by an atomic absorption spectrometry. The remaining fraction was heated till benzene was removed completely. The precipitate of beryllium hydroxide was made with ammonia water and was ignited at 750°C to convert it to BeO in an electric furnace. The γ -activity of BeO including Be-7 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% as a mean).

The particles obtained with sediment traps were filtered with glass fiber filters (Whatman GF/C) or Nucleopore filters (0.6 microns in pore size). Followed by the digestion of the samples on filters by immersing them into a mixture of HF, HNO_3 and HClO_4 , one twentieth fraction of the solution was analyzed for Th-234 and the remainder was used for the analyses of Be-7, Th-228, Th-230 and Th-232. The subsequent analytical procedures for Th-234 and Be-7 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. Their alpha and beta activities were counted.

II - 2 Sediment traps employed for this research

a) U-type trap

This type trap was a relatively small one designed for the easy operation on small vessels. This was made with an poly-vinyl chloride pipe (60 cm length, 7.5 cm diameter). The 4 - 6 pipes were knoted together and moored at several depths for a few days. The observations of particulate flux were carried out using this type traps once a month or more times. The design of this trap and the typical mooring configuration operated in Funka Bay are shown in Figure II - 2.

b) T-type trap

T-type sediment trap was designed for the collection of a large amount of settling matter during a short period. This trap consists of twin rectangular P.V.C. boxes and each opening area and height are 0.67 m^2 and 25 cm, respectively. A pyramidal P.V.C. funnel (30 cm height) and a plastic bottle are connected to the each box. Figure II - 3 shows the design with two photographs and the mooring configuration of this trap is illustrated in Figure II - 4. The trap experiments with this trap were carried out only in Funka Bay in 1977 and 1978, since the many difficulties at the operation on the vessel arose from its too large size. Although the trap had a lid to avoid the loss of sampled material at the retrieval operation, the lid did not work well.

c) H(NH)-type trap

For the collection of settling material in open sea, H(NH)-type trap was devised. This trap is illustrated in Figure II - 5, which is composed of 6 cylinders for a collector and a frame made of stainless steel. Two types of the cylinders were used, which were "H-type trap" (Figure II - 6) and "NH-type trap" (Figure II - 7). Each cylinder had a lid which prevented the loss of collected material at the retrieval operation. The mouth of each cylinder was covered with a net (1 cm in mesh size) to protect the invasion of zooplankton and neuston. In shallow water, NaCl and NaNO₂ were used for an antiseptic, but any antiseptic was not used in deep water of the open ocean, since the decomposition of trapped matter was not expected to occur significantly because biological activity in the deep water might be low as discussed by Honjo et al. (1978).

I - 3 Results and discussion

II - 3 - 1 Be-7, Th-234, Po-210 and Pb-210 in seawater.

The vertical profiles of Be-7 and the density of water (σ_t) in Funka Bay are plotted in Figure II - 8. The profiles of σ_t show that the seasonal thermocline developed well in July and August in the surface 40 m layer, but it was not obvious in May. The overall analytical error for Be-7 is not considered to be larger than the counting error of about 11 % by referring the results of duplicate analyses of the same sample. The concentration of Be-7 in May was relatively constant with depth, showing 240 dpm/1000l in the surface water and 184 dpm/1000l in the water of 70 m depth. However, the concentration

of Be-7 in the intermediate water increased with time from July to August and a maximum value was always found in the layer. The maximum of Be-7 in this Bay in summer is difficult to be explained by means of mixing with water outside the Bay and/or the fluctuation of the input rate of Be-7 from the atmosphere. The particulate transport of Be-7 seems to be responsible for its vertical profile, although Silker et al. (1973) pointed out that the vertical profile of Be-7 in the surface 100 m of open sea is not influenced by the particulate transport.

Th-234, Po-210 and Pb-210 in seawater were also determined at the same site on Aug. 20, 1979 and the results are given in Figure II - 9 together with the density of seawater (σ_t). The profile of σ_t on Aug. 20, 1979 was almost the same as that on Aug. 22, 1977. It is interesting to note that all these nuclides were concentrated in the same intermediate layer. Many previous studies on these nuclides have not observed such a clearly concentrated layer in the coastal sea. The coastal sea may be too shallow or the sampling depth intervals may be too coarse to find maxima in their studies. However, we can find maximum of Th-234 in the vertical profile of Minagawa and Tsunogai (1980) who rather crudely determined Th-234 in seawater at stations near St. P in this study on Aug. 12, 1974.

Th-234 is one of radioisotopes produced in seawater, but a half of Po-210 a substantial part of Pb-210 and all of Be-7 in the surface seawater are air-borne. On account of the difference in their source functions, the same shape in vertical profiles of these nuclides strongly suggested that these nuclides are transported from the surface to the depths by the same

conveyer and regenerated or dispersed after broken to small particles in the intermediate layer. It seems likely that the incorporation of these radioisotopes into organic particles is intimately related to their sinking from the surface to the intermediate layer where the decomposition of organic matter is actively occurring as discussed by Tsunogai and Minagawa (1978). However, the decrease of the dissolved oxygen contents in the layer was not so large (Figure II - 9b) and the regenerated amounts of the radionuclides estimated from those of nutrients and the concentration ratios of these nuclides to the nutrients in settling particles in seawater were insufficient to explain the surplus in the intermediate layer. This may indicate that the settling particles were broken down to small particles of negligibly small sinking velocity in the layer.

II - 3 - 2 Be-7 and other nuclides in settling matter.

To find a direct evidence of the particulate transport of Be-7, sediment traps were deployed three times in the water of 40 m and 80 m depths at St. P in 1977. The specific activities of Be-7 were nearly constant with depth. This fact is to be noted, because Be-7 of surface origin could be expected to decrease with increasing depth if the particles containing Be-7 sank with a definite velocity. The possible explanations for the fact are as follows;

1. A sufficiently large sinking velocity of the settling particles such as fecal pellets. This means that all the particles sinking seawater have the virtually same young age.

2. Resuspension of the bottom sediments by the active convection of water which is caused by the cooling of water in winter.
3. Cancellation of two opposite effects - the reduction of particles by the dissolution or the decomposition which relatively concentrates insoluble Be-7 and the radioactive decay out of Be-7 which decreases its concentration in particles.
4. Ceaseless trapping of these nuclides into the settling particles from seawater, especially in the deep layer where the sinking velocity slows down. This means the concentration of the nuclide in the particles increases with time before it appreciably decays out.
5. Rather repeated transformation of particle sizes by aggregation between large particles which can settle down and colloidal particles. Consequently, particulate matter has sufficiently high sinking velocity as a whole. This mechanism is the same as Tsunogai and Minagawa's settling model (1978).

The fact can not be explained by the first possibility alone because the radioactive disequilibrium between U-238 and Th-234 was more remarkable in the deep water (Figure II -9a), which needs active removal in the layer and because the maximum of Be-7 has been found in the intermediate water as mentioned above, which needs its regeneration in the layer. The second possibility is also deniable, because the Bay water in summer was well stratified. Furthermore, we know a fact that Th isotopes were more concentrated in settling particles collected at 80 m depth than those at 40 m depth. This fact is favorable for the fourth

(Figure II -12)

and the fifth possibilities. For further discussion about the possibilities, the activity ratios of Be-7/Th-234, Th-228/Th-234, Th-230/Th-234 and Th-232/Th-234 of the sediment trap samples were calculated (Table II - 2). The Be-7/Th-234 ratios at 40 m depth were higher than those of 80 m depth in the summer of 1977. This fact can be explained by the fourth or the fifth possibility, namely by a mechanism that sinking particles continuously pick up Th-234 or interchange these nuclides with those in seawater. If the interchange does not occur, Be-7/Th-234 in sinking particles should increase with depth, because the half-life of Be-7 is longer than that of Th-234. Third possibility also needs a constant Be-7/Th-234 ratio or the ratio increasing with depth. The small Be-7/Th-234 ratio observed at 40 m depth in autumn may be due to the Tsugaru Warm Current flowing into the Bay in that season. The higher specific activities of Th-230 and Th-228 at 80 m depth may be an evidence for the validity of the fourth or the fifth possibility, because the specific activities of Th-232 observed at 80 m depth was not higher than those of Th-232 observed at 40 m depth, except the result observed during the period from July 26 to Aug. 22. Finally, we suggest the fifth possibility to be the most favorable explanation for the mechanism of particulate removal of these nuclides from seawater, because the most of these nuclides may be already incorporated into smaller suspended particles before larger settling particles pick up them. Besides the facts given above, these nuclides are concentrated in the intermediate water as shown in the proceeding section. These facts contradict with the irreversible removal of these nuclides from seawater into sinking particles.

II - 3 - 3 Net fluxes of Th-234 and Be-7

The net removal of Th-234 estimated from its deficiency from U-238 in sea water observed by Minagawa and Tsunogai (1980) and the flux observed with sediment traps are compared in Table II - 3. The observed fluxes of Th-234 with sediment traps were about ten times smaller than those of the estimated. This small trapping efficiency may be caused by the shape of the trap under the relatively high current velocity of coastal sea water as discussed by Gerdner (1980) and the loss during the retrieval operation. Assuming the Be-7/Th-234 ratio (R) of material collected with the sediment trap 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 sea water as follows;

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

Where A_{Be} and A_{Th} are the inventories of Be-7 and Th-234 (dpm/m²), respectively, and τ_{Th} is the residence time of Th-234 (days) which has been obtained by Minagawa and Tsunogai (1980).

The results calculated are also given in Table II - 3. The fluxes of Be-7 observed with the sediment traps were also about ten times smaller than the calculated ones of Th-234. The residence time of Be-7 in the water column of 80 m depth is estimated to be 120 - 160 days, which is somewhat longer than that of Th-234. However, the above estimation includes rather large uncertainties and then the difference between the residence times of the two nuclides may not be significant. Therefore we can

conclude that Be is one of the most active elements for the removal from seawater like Th-234.

In the previous studies (Silker et al., 1973), Be-7 was used for an estimate of the eddy diffusivity of the surface sea water as a radioactive tracer with the same way as Tritium. However, the obtained vertical diffusivity may not be real unless the effect of particulate transport is taken into consideration. The fact of active removal of Be-7 as well as Th-234 in the surface layer of the ocean also shows us its usefulness to study the mechanisms of particulate removal of metals in sea water and of the downward transport of particles from the surface to the deep.

II - 3 - 4 Total particulate flux observed with U-type sediment traps and total suspended matter in sea-water.

Total particulate fluxes observed with U-type sediment traps at St. P from 1977 to 1980 are given in Figure II - 10 and II - 11. The traps were deployed at the water depths of 1 m, 40 m, 60 m and 80 m. The fluxes observed at 1 m, 40 m and 80 m depths are shown in the figures together with total suspended matter observed at the same depths. It should be noted that the total particulate fluxes increased in autumn and winter of 1977 and the fluxes observed in December was about twenty times higher than those observed in September. The sedimentation rate of the bottom sediment at the same site has been estimated to be $2.7 \text{ mg/m}^2\text{day}$ by the Pb-210 dating method (Tsunogai et al., 1977: see Appendix I). The observed flux in winter much exceeded the sedimentation rate and it should be noted that the concentration of total suspended matter in sea water did not so increase in winter. This enormous particulate flux in winter season has been observed in every year (Figure II - 11). The large particulate flux observed with sediment traps in every winter season may be caused by one of the following reasons.

1. Resuspension of the bottom sediments by the active convection of the water, which is caused by cooling of the water.
2. Temporally high sedimentation in winter. The strong winter monsoon may bring the continental dust to this Bay in winter season.
3. Apparent high sedimentation caused by the repeated transformation of particle sizes between large aggregate particles which can settle down and colloidal particles which move up convective water. This process has already mentioned in section II - 3 - 2.

4. Erroneous observed sedimentation due to the design of the sediment trap used.

The second possibility is considered to be impossible. We can not find a large source of particulate matter to the Bay in this season. We do not know a fact that the river input increases in winter. The input rate of atmospheric dust does not increase in winter (Shinagawa, 1980). Although the biological production of particles is also a source of the particulate flux, the bloom of plankton does not occur in winter (Hirakawa, 1976).

II - 3 - 5 "Upward flux" of particulate material in Funka Bay.

The specific activities and fluxes of Th-234 observed with U-type sediment traps provide an information on the source of the particles collected (Figure II -12). The specific activities of Th-234 were approximately constant, although the total particulate fluxes increased extremely. This implies that the large flux observed in winter was not due to the resuspension of the bottom sediment. If the resuspended matter from the sediment was trapped, the specific activity of Th-234 of trapped matter in winter should be lower than that in summer because of the low concentration of Th-234 in the surface sediment (Santshi et al., 1979). Thus, the first possibility can be deleted. I propose the third possibility for the mechanism because the flux observed in winter is too large to be explained by the particles sinking downward unilaterally. The observed fluxes of Th-234 with sediment traps in winter (Figure II - 12) much exceed the net removal flux obtained from the disequilibrium between U-238 and Th-234 in sea water (Table II - 3) and it is

noted that the concentration of Th-234 in sea water did not decrease with the inordinate large flux (Figure II - 13).

This means that the fluxes observed with sediment traps do not give the net removal flux from sea water. The sediment traps are considered to catch the particles which sink downward in sea water. It needs the occurrence of the upward particulate flow in the water column against its gravitational settling. Thus, the surplus flux observed in winter may be due to the active convection which moves up small particles together with water and the settling of aggregated large particles (Figure II - 14).

Is this a specific phenomenon in Funka Bay? I have carried out the same sediment trap experiments and determined total suspended matter and radioisotopes in sea water in the summer of 1978 and in the winter of 1980 in Tokyo Bay.

II - 3 - 6 "Upward flux" of particulate material in Tokyo Bay.

The sampling site and the trap system employed in Tokyo Bay are shown in Figure II - 15 and II - 16, respectively. The H(NH) and U-type traps were used for the collection of sinking particles. The flux in winter observed was higher than that observed in summer. However the difference of the total particulate flux between the two seasons was smaller than that observed in Funka Bay. It may be due to the active biological production of particulate material in the summer of Tokyo Bay, which is famous for a ^{very} extremely polluted area. The data of ignition loss show high contents of organic material in the trapped matter in summer (Table II - 4). The specific activity of Th-234

observed in summer was lower than that observed in winter. It is also a result of the dilution with the biogenic matter.

The density of water (σ_t) shows the existence of stratified water in summer and that of mixed water in winter (Figure II - 18). The concentration of Th-234 in sea water was nearly equal to that observed in winter (Figure II - 19). However, the activity ratio of Th-234/U-238 was around 0.30 in both seasons, which was about half of that obtained in Funka Bay. The net removal flux of Th-234 can be obtained from the disequilibrium between U-238 and Th-234 in sea water, assuming a steady state condition (Table II - 5). The flux of Th-234 observed with sediment traps in summer was about equal to the net flux calculated but that observed in winter exceeded the net flux.

Almost all the observations carried out, except the blooms of plankton in summer season, gave the same results as those obtained in Funka Bay. The concept of "Upward flux" of particulate matter seems to be also true in Tokyo Bay.

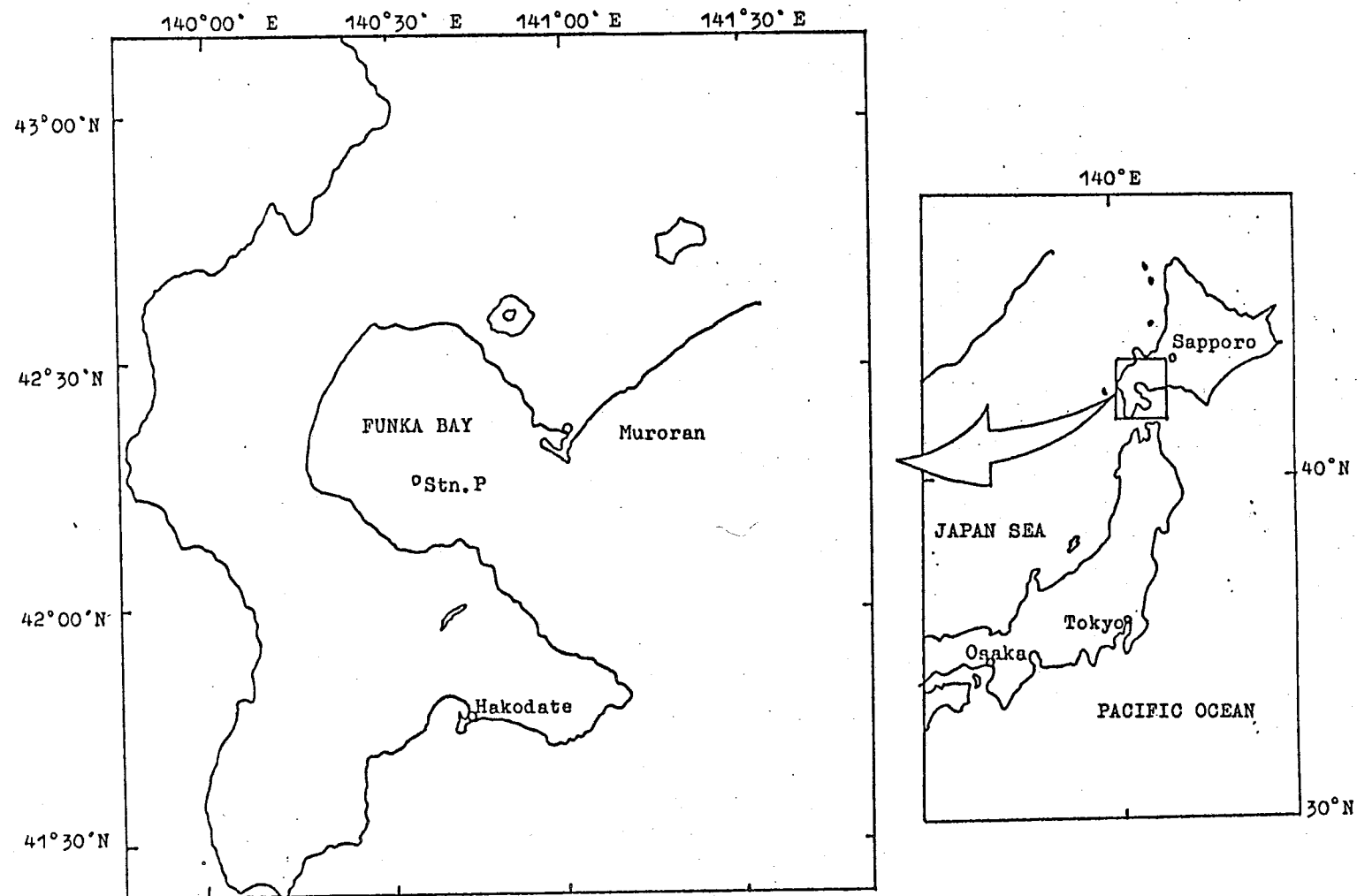


Figure II -1 Sampling sites in Funka Bay.

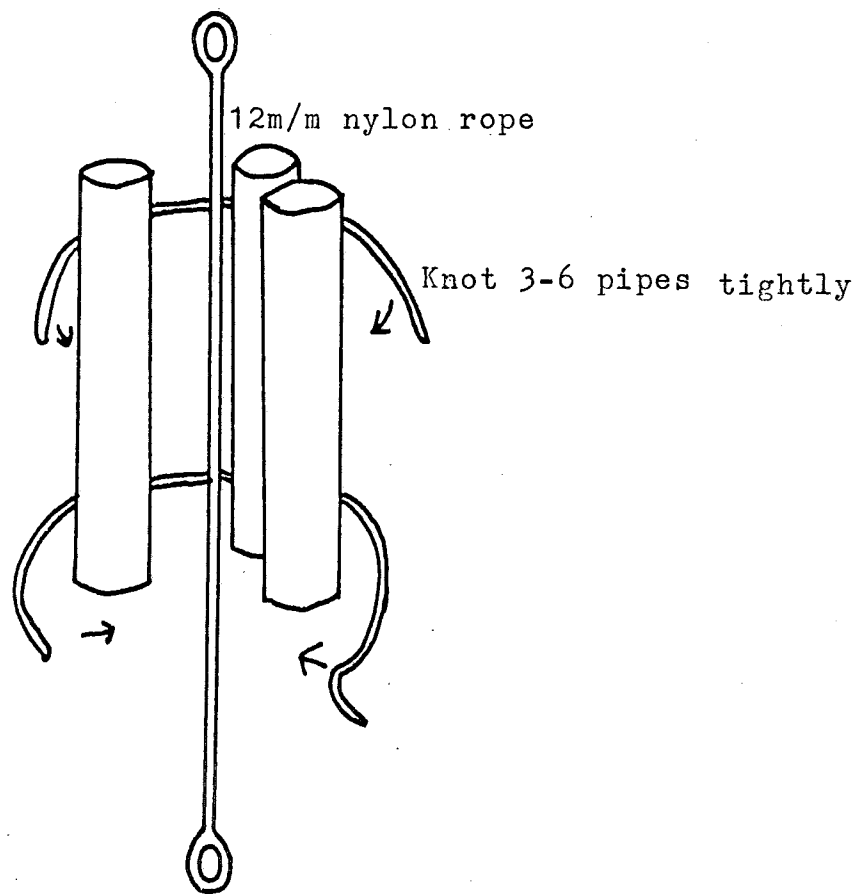
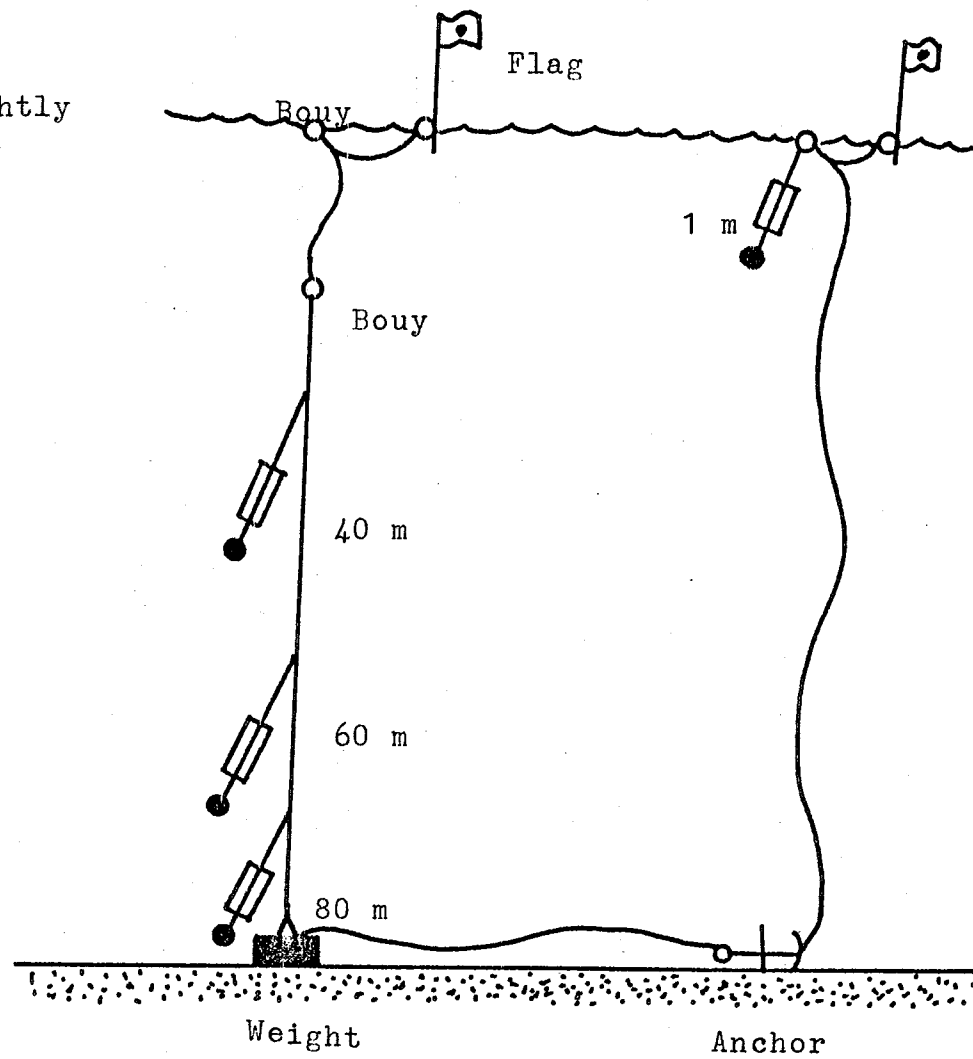


Figure II - 2
U type trap and its mooring condition.



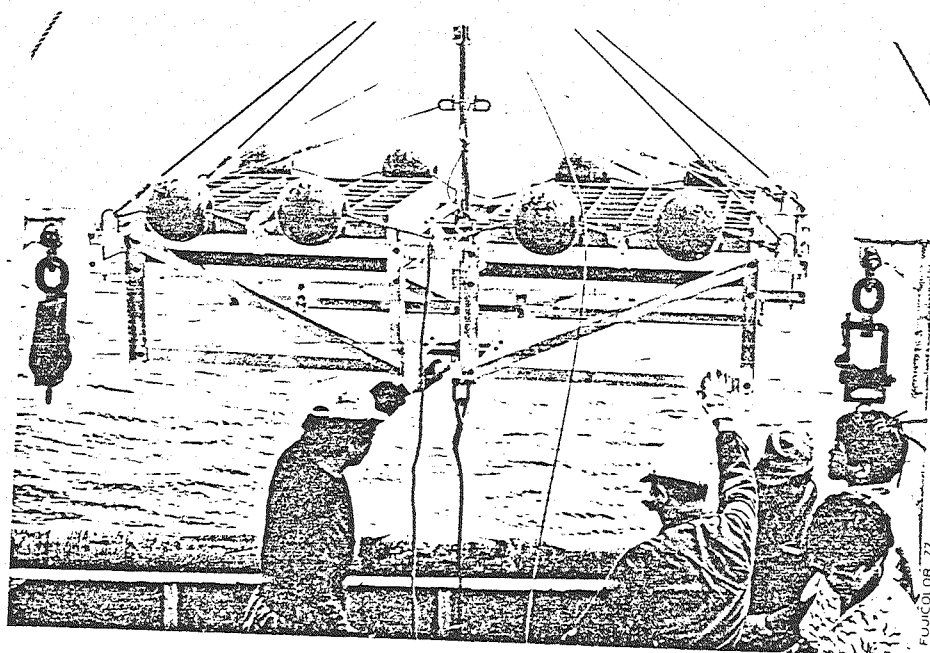
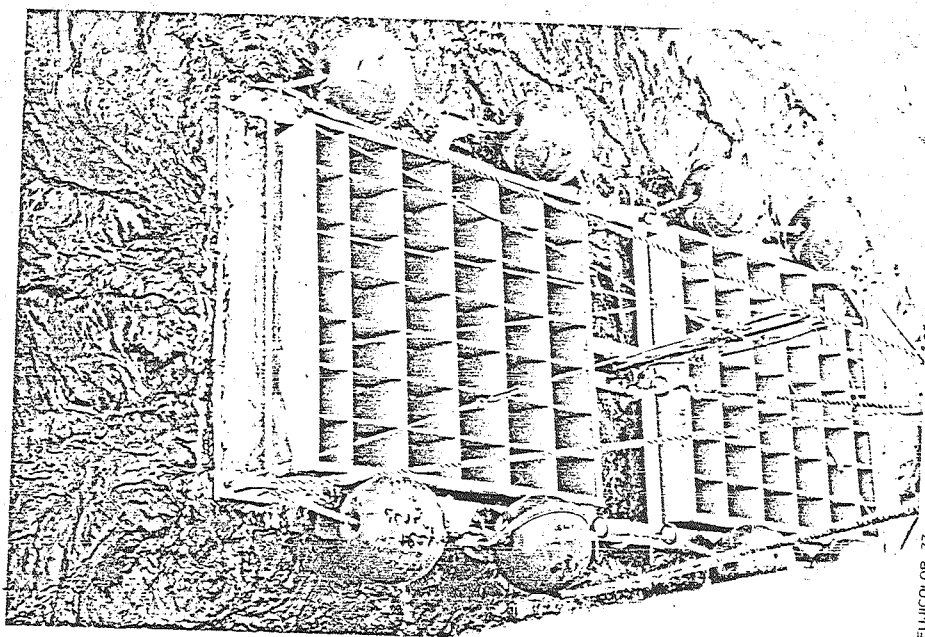


Figure II - 3. Photographs of T type sediment traps.

Figure II -4. Mooring condition of T type sediment traps at St. P in Funka Bay.

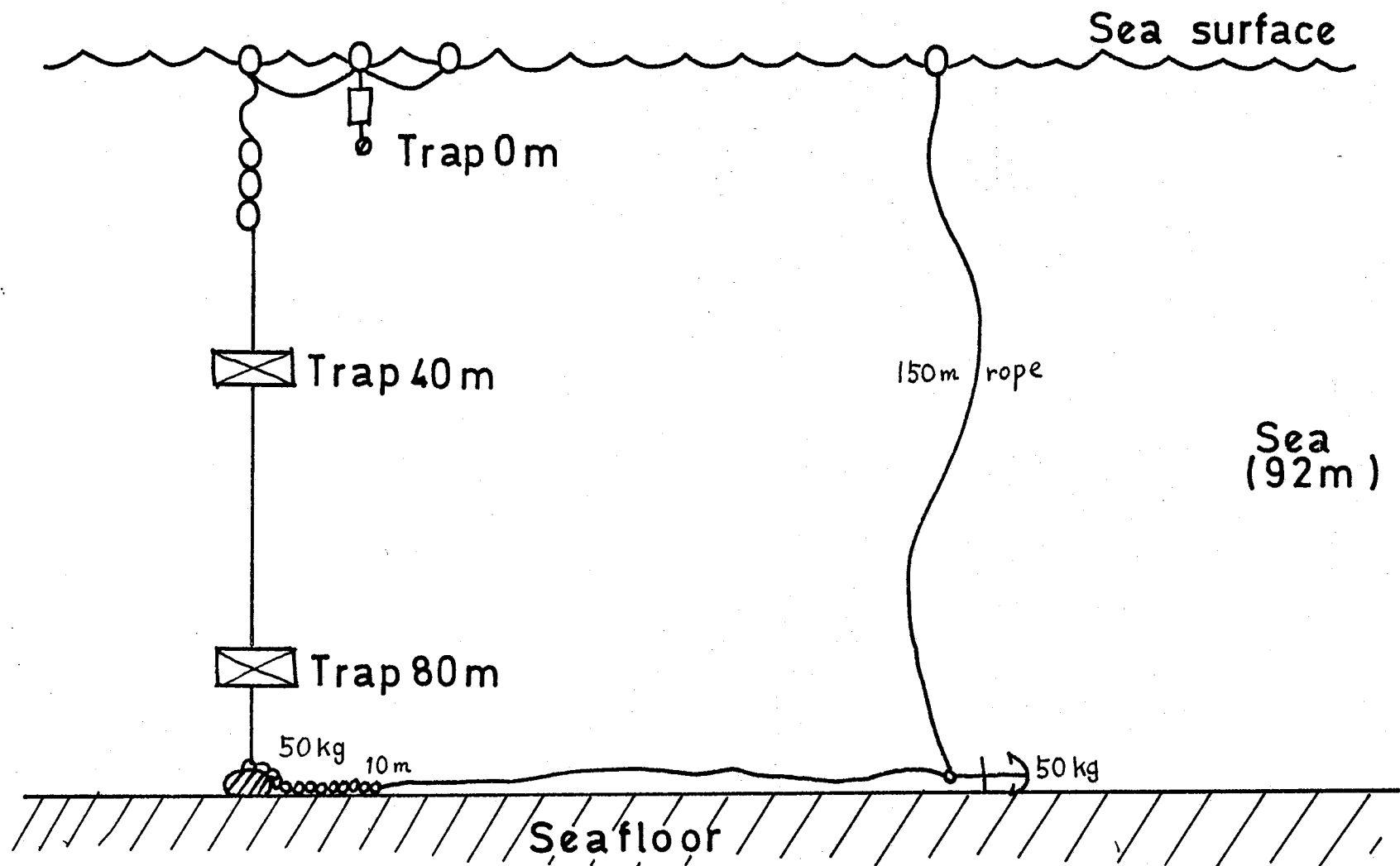


Figure II -5. Configuration of H(NH) type sediment trap

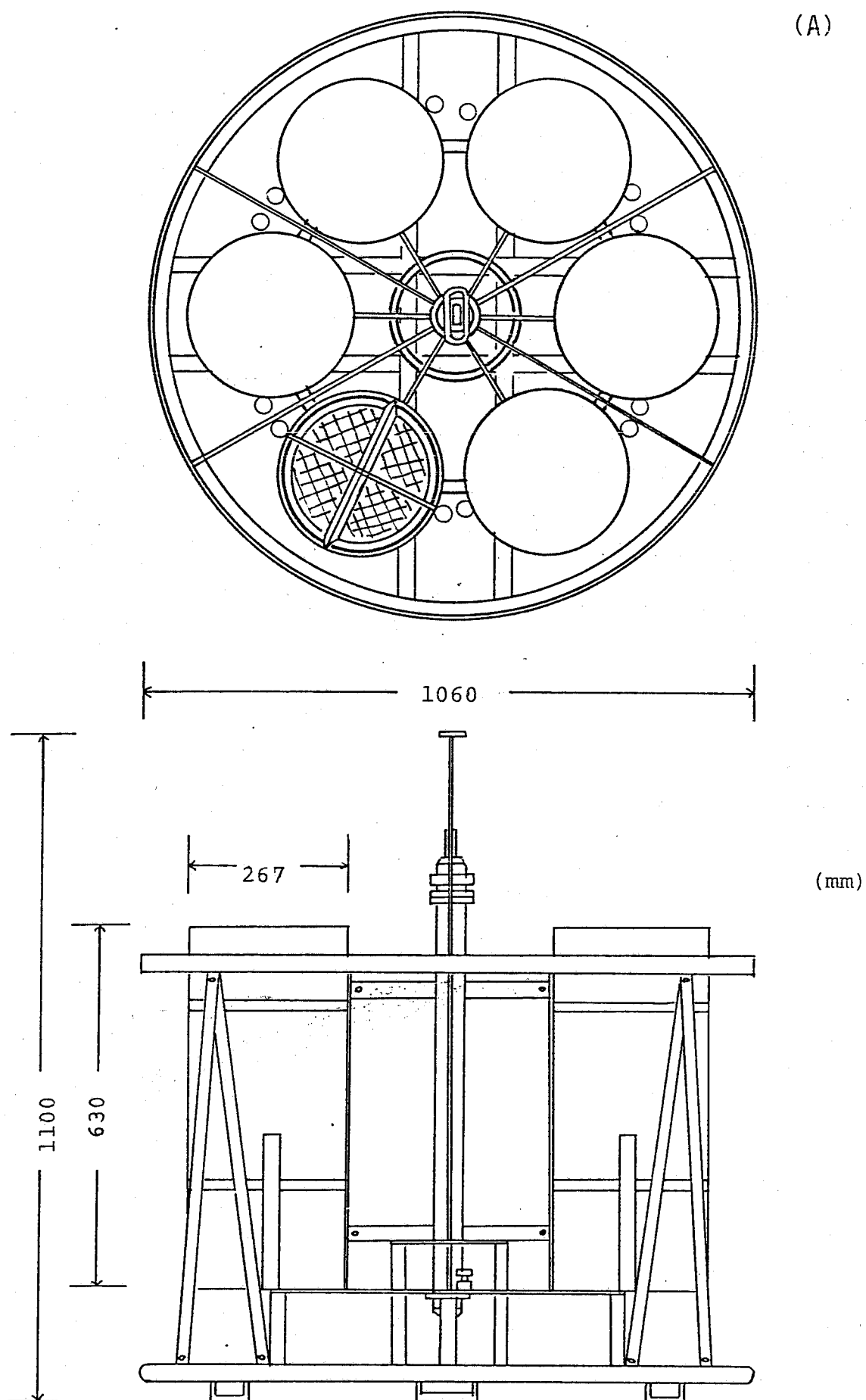


Figure II -6.

Structure of H type collector..

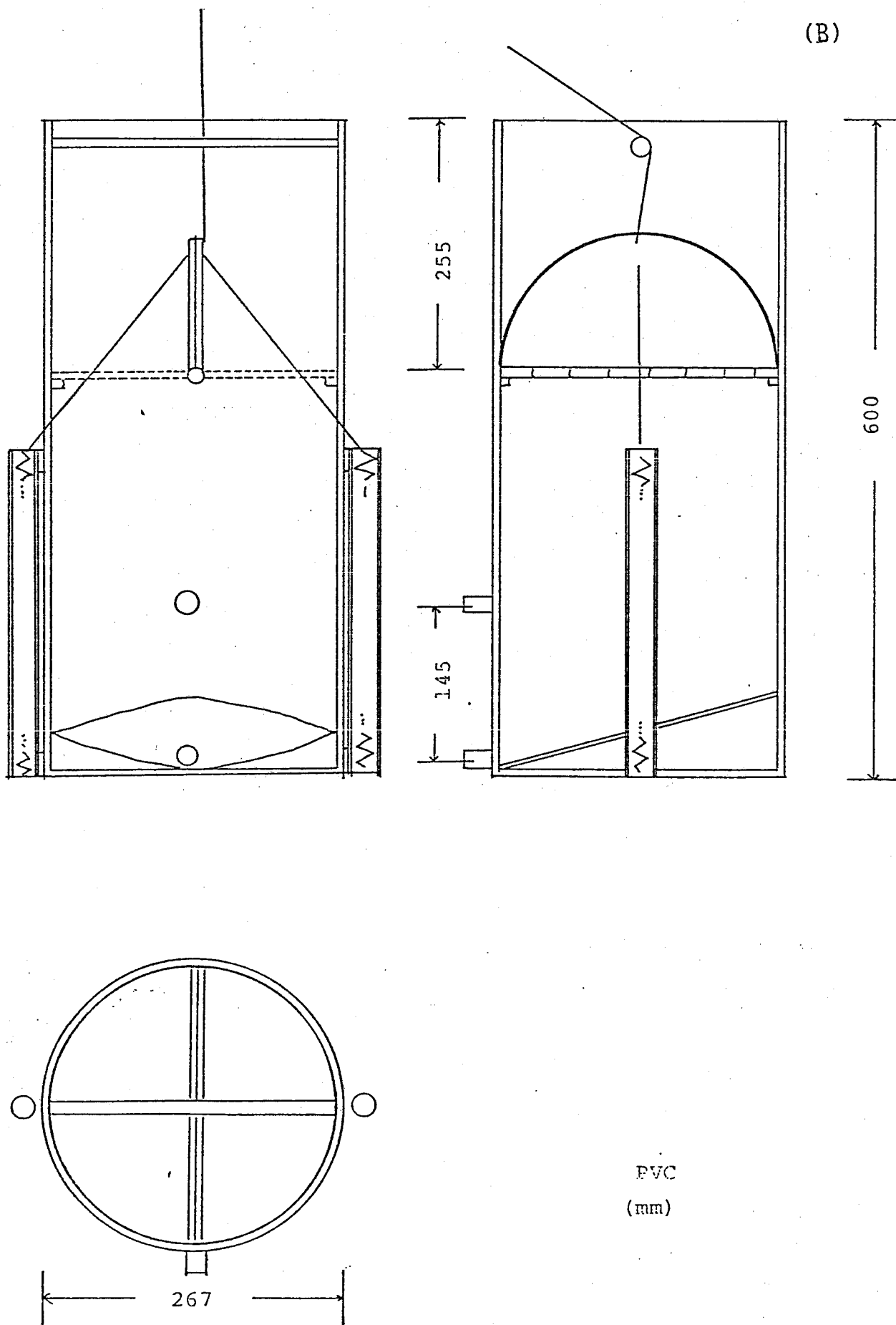
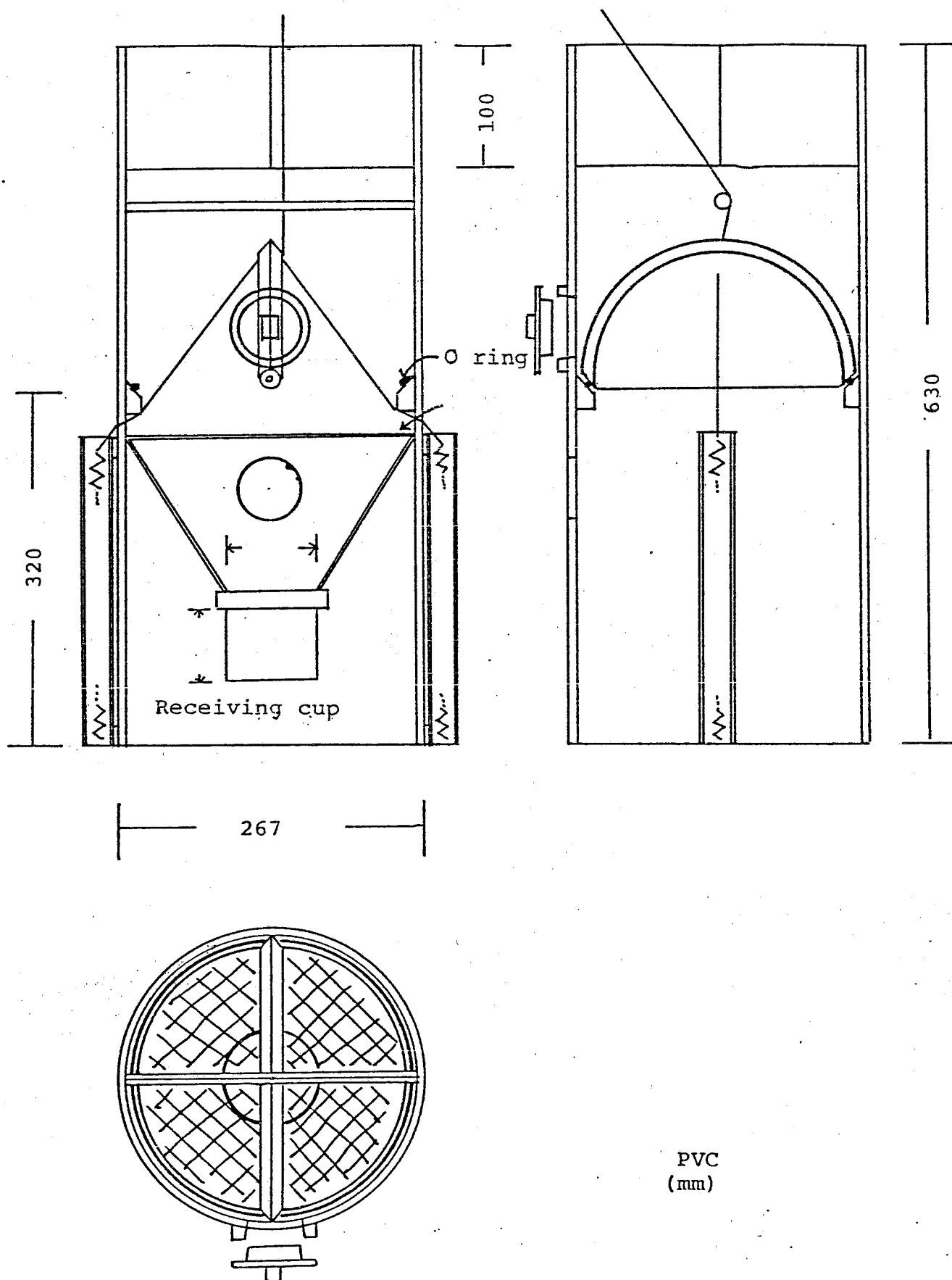


Figure II -7.

Structure of NH type collector.

(B)



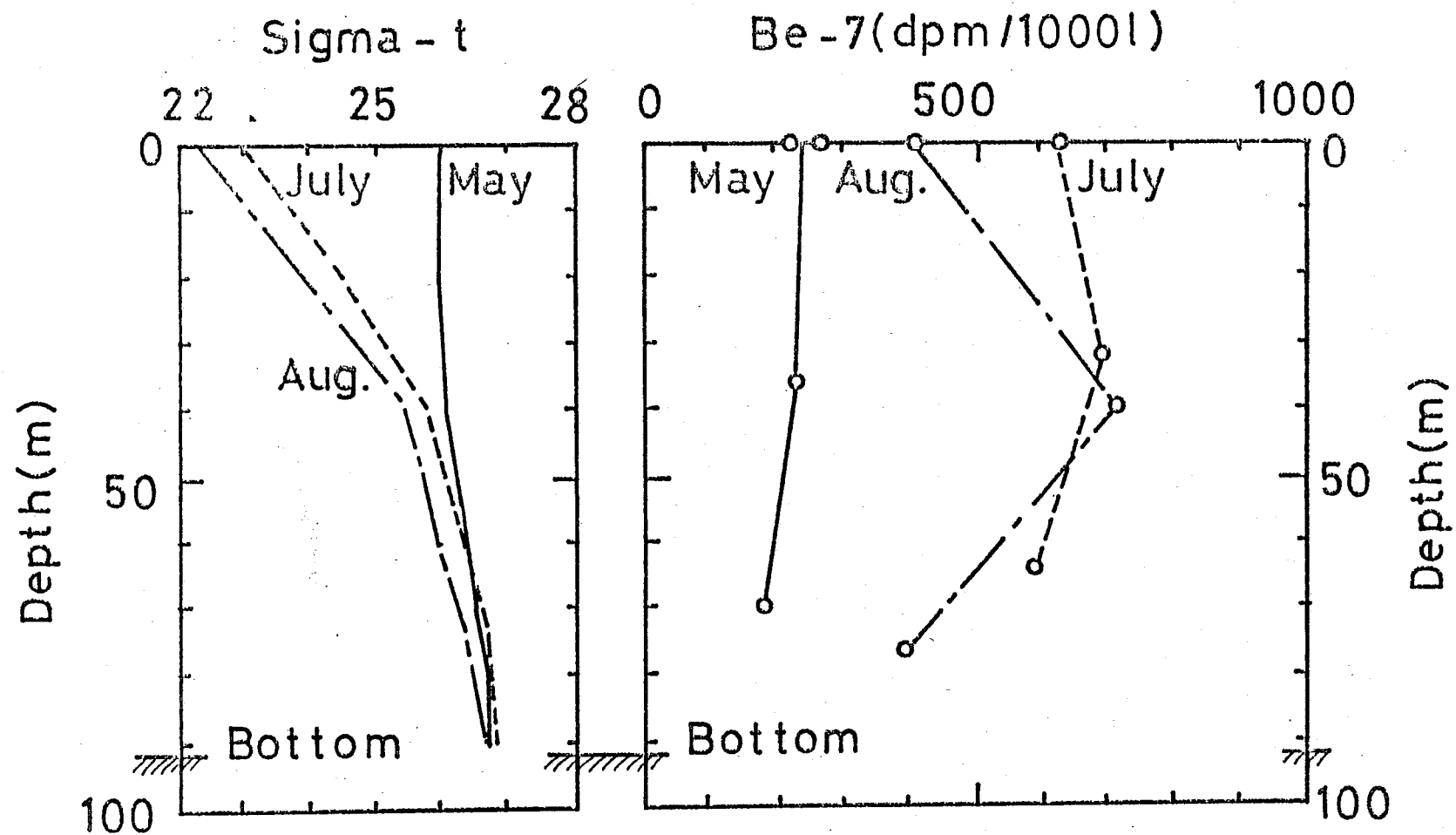


Figure II -8. Vertical distribution of Be-7 in sea water and sigma-t at St. P in Funka Bay in the summer of 1977.

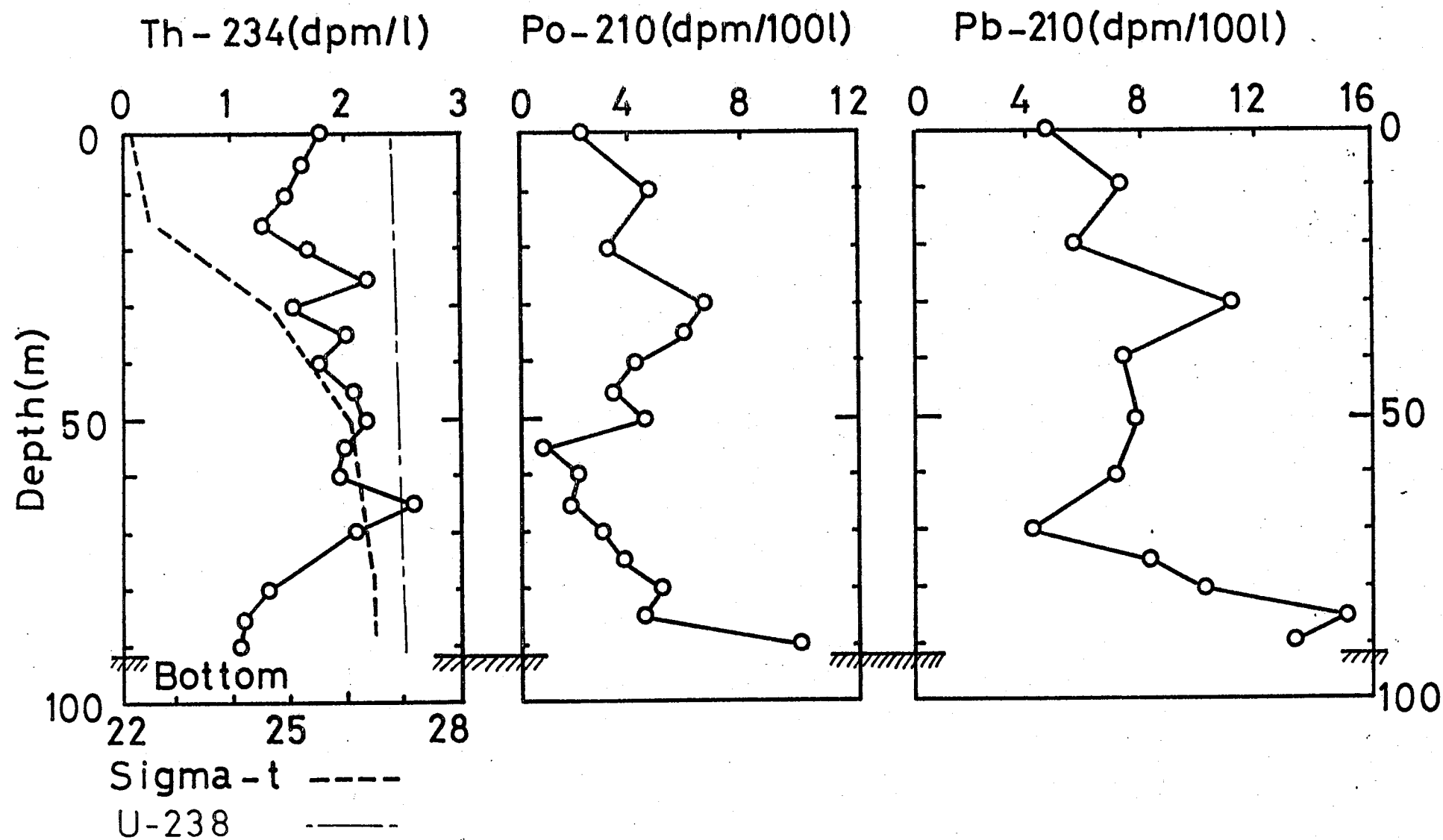


Figure II - 9a Vertical distributions of Th-234, Po-210, and Pb-210 in seawater at St. P in Funka Bay on Aug. 20, 1979.

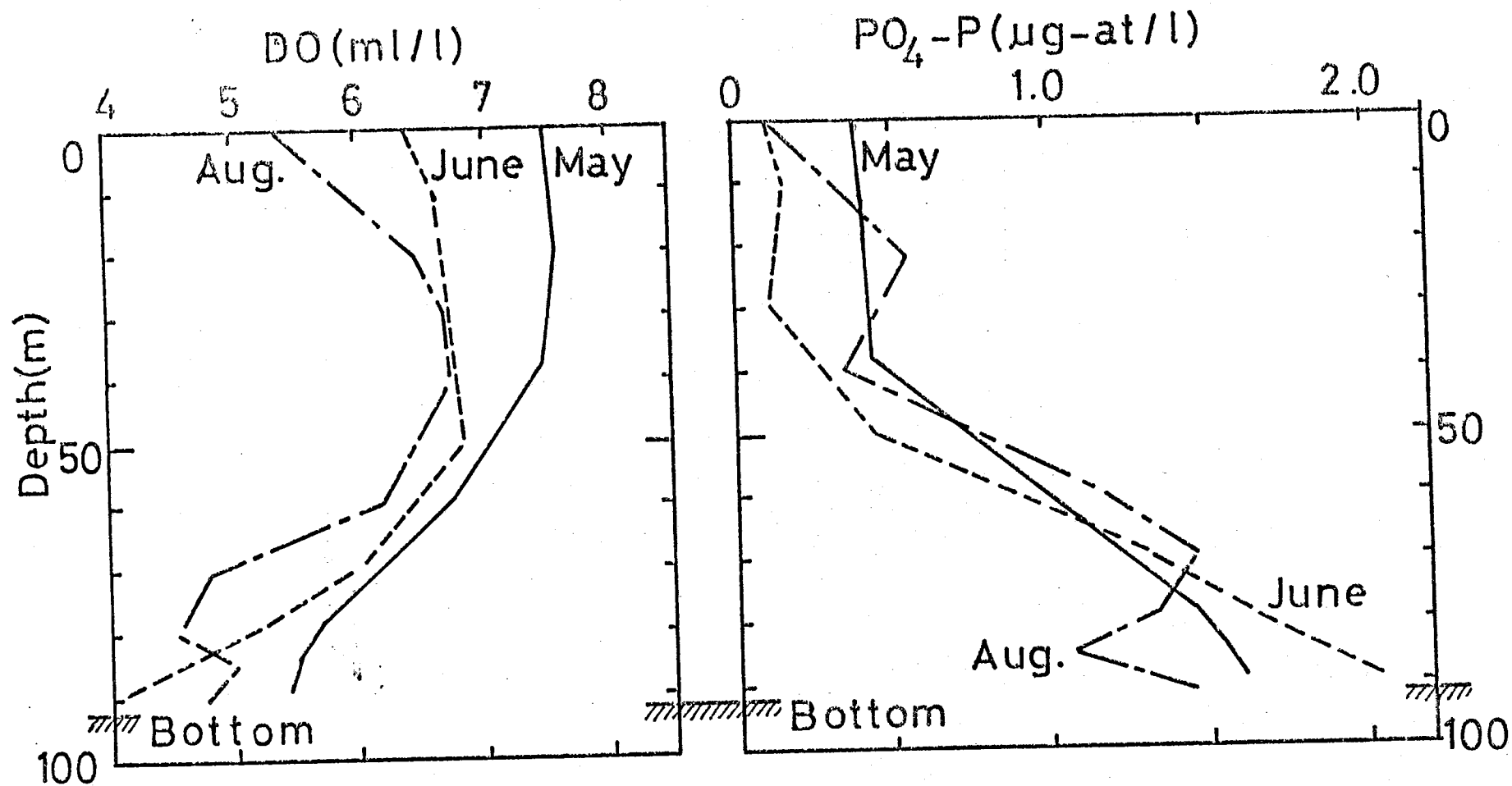


Figure II - 9b Seasonal variations of vertical distributions of dissolved oxygen and reactive phosphate at St. P in Funka Bay in the summer of 1977.

Figure II -10. Total particulate flux observed with U type sediment traps and total suspended matter in sea water.

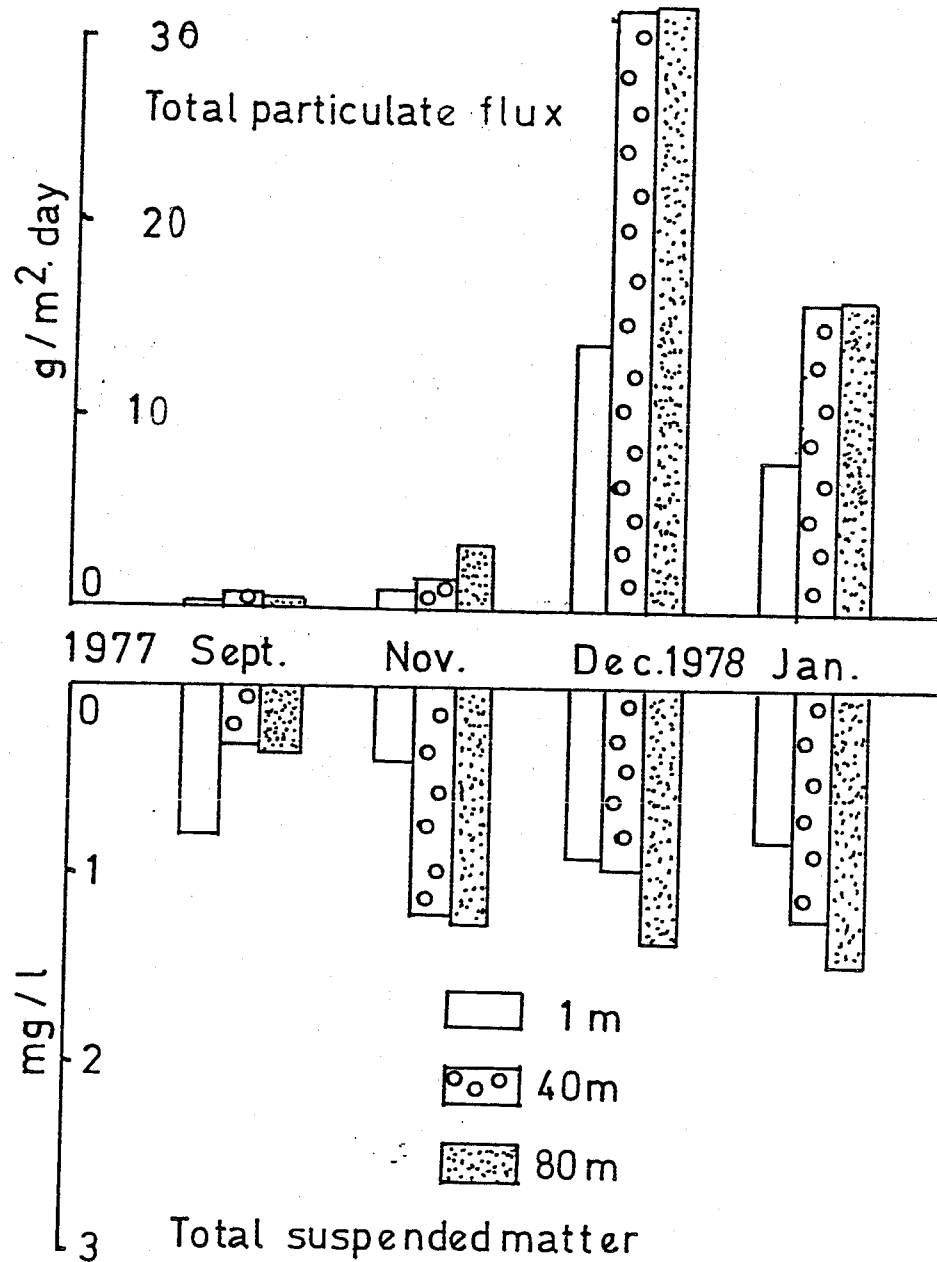


Figure II - 11. Total particulate flux observed with U type sediment traps and total particulate matter in sea water from Jan., 1978 to Mar., 1980.

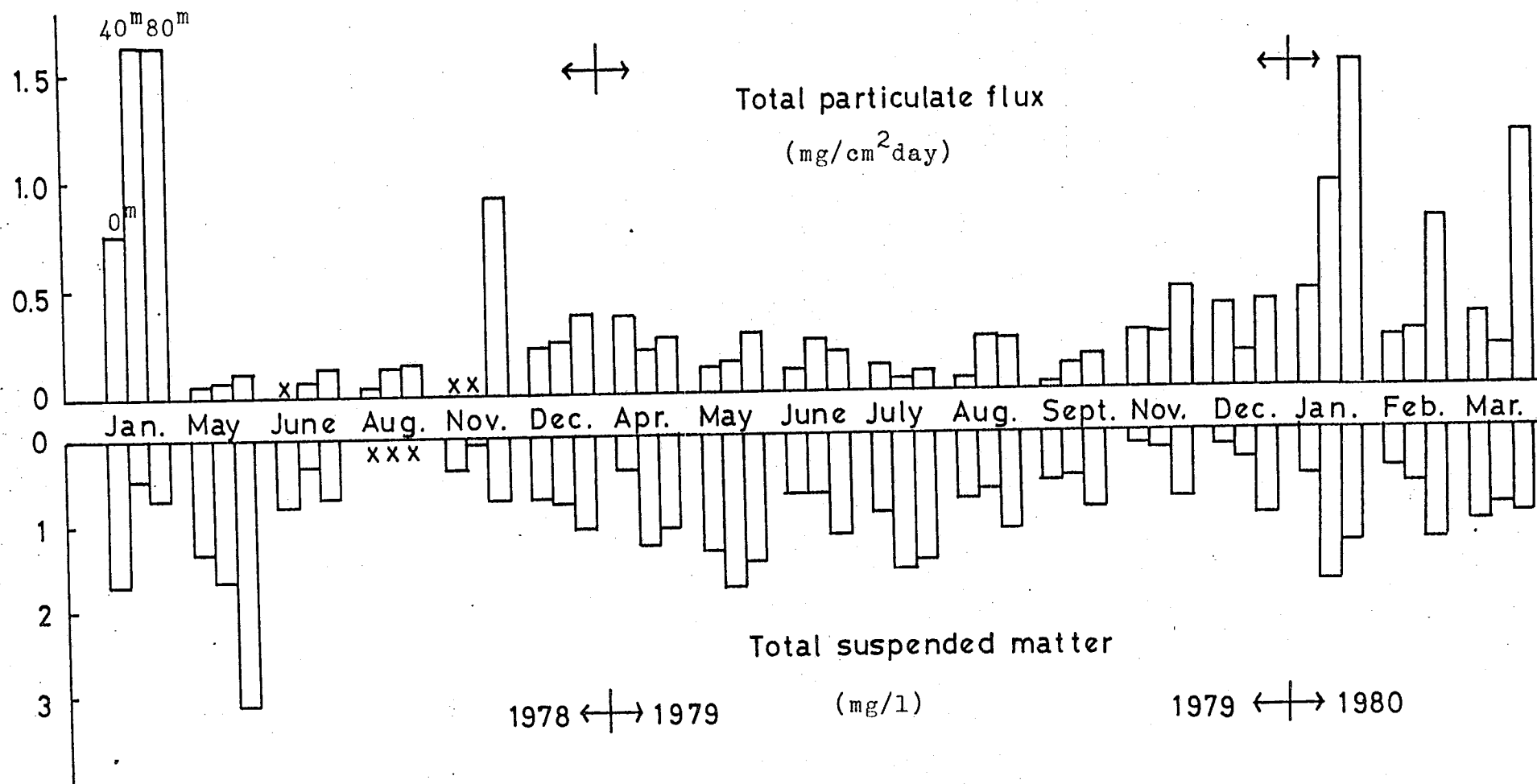
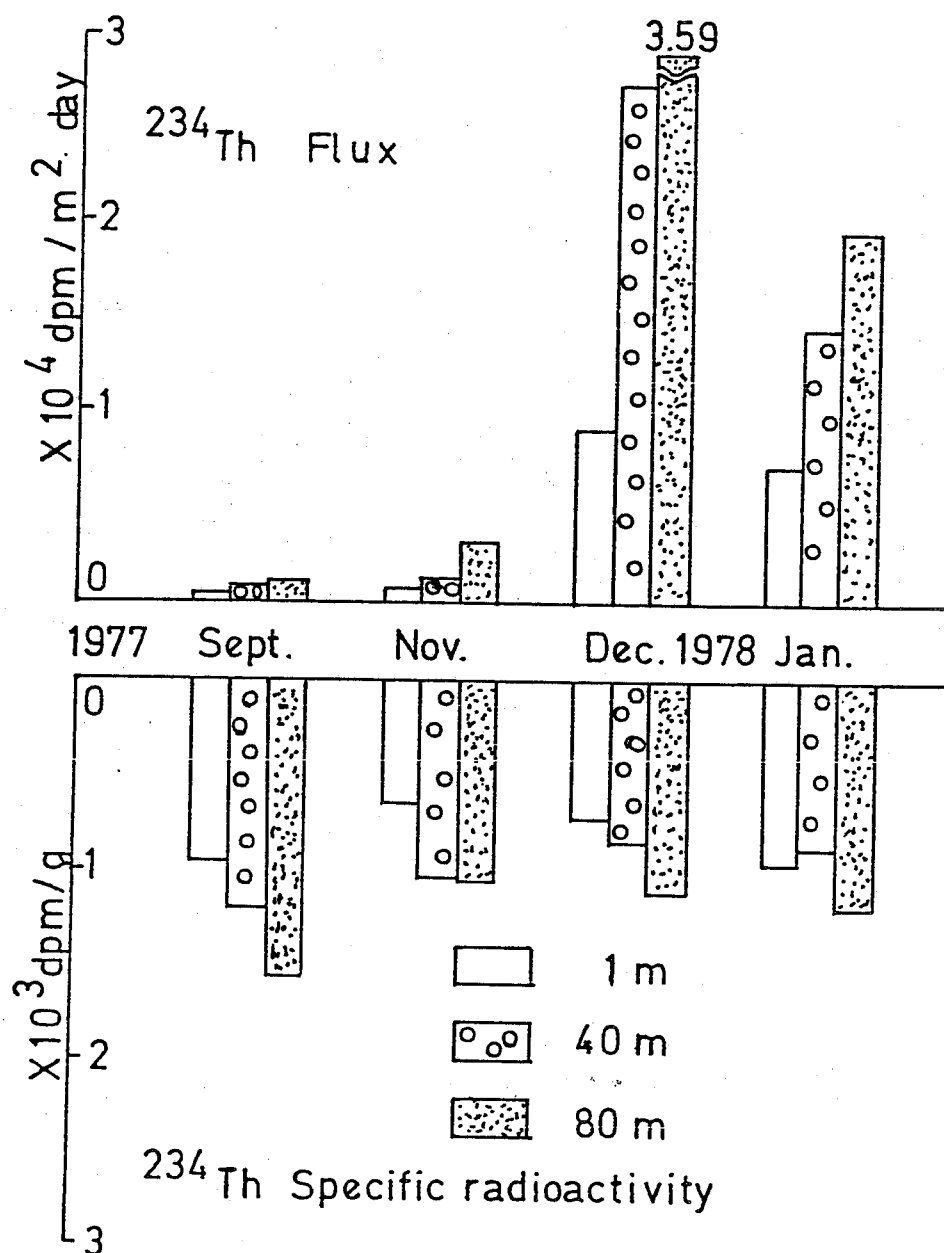


Figure II -12. Specific activities of Th-234 in settling matter collected with U type sediment traps and fluxes of Th-234.



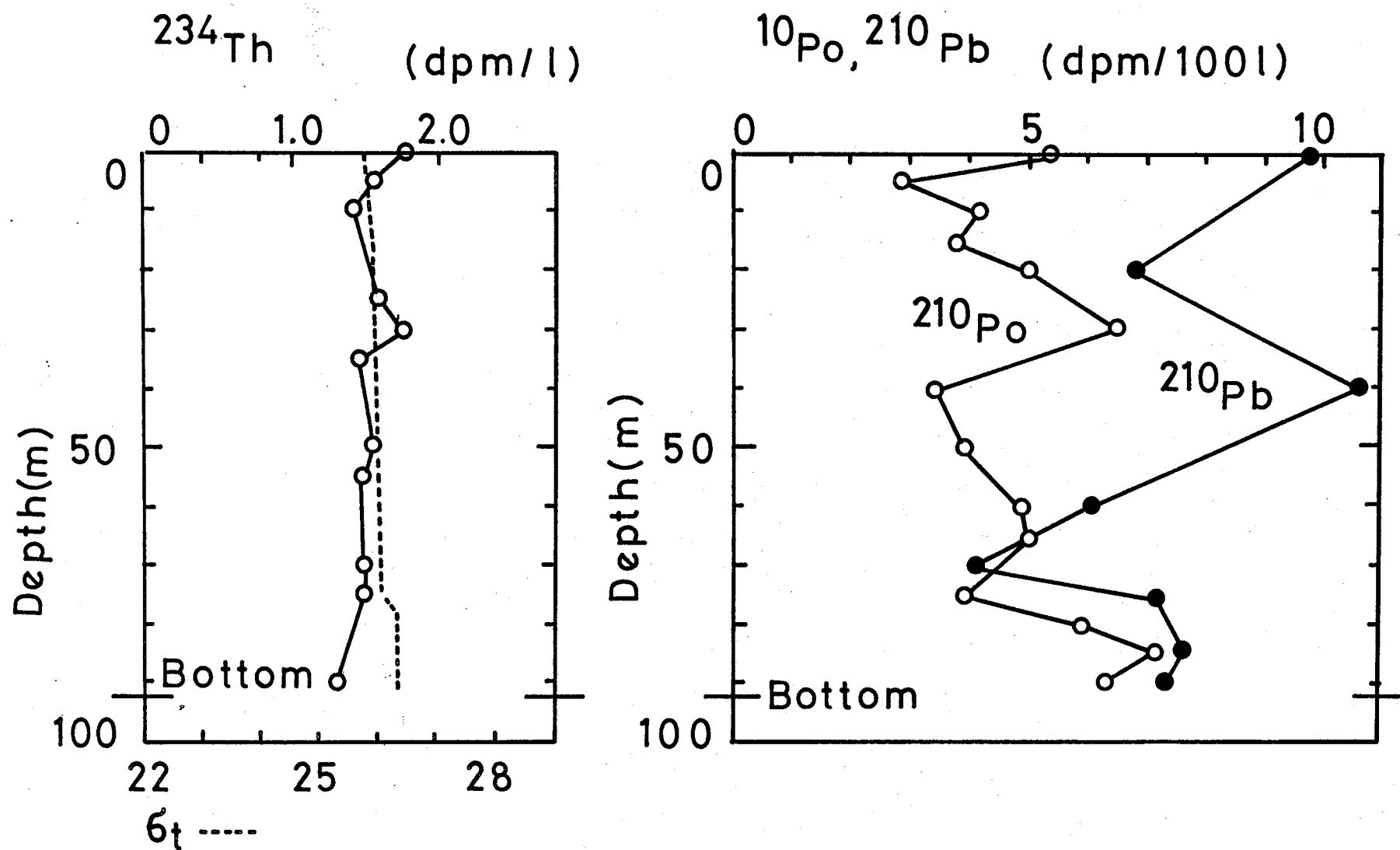


Figure II - 13 Vertical distribution of Th-234, Po-210, and Pb-210 in seawater on Dec. 12, 1979.

Figure II - 14. Seasonal variation of water density at St. P in Funka Bay.

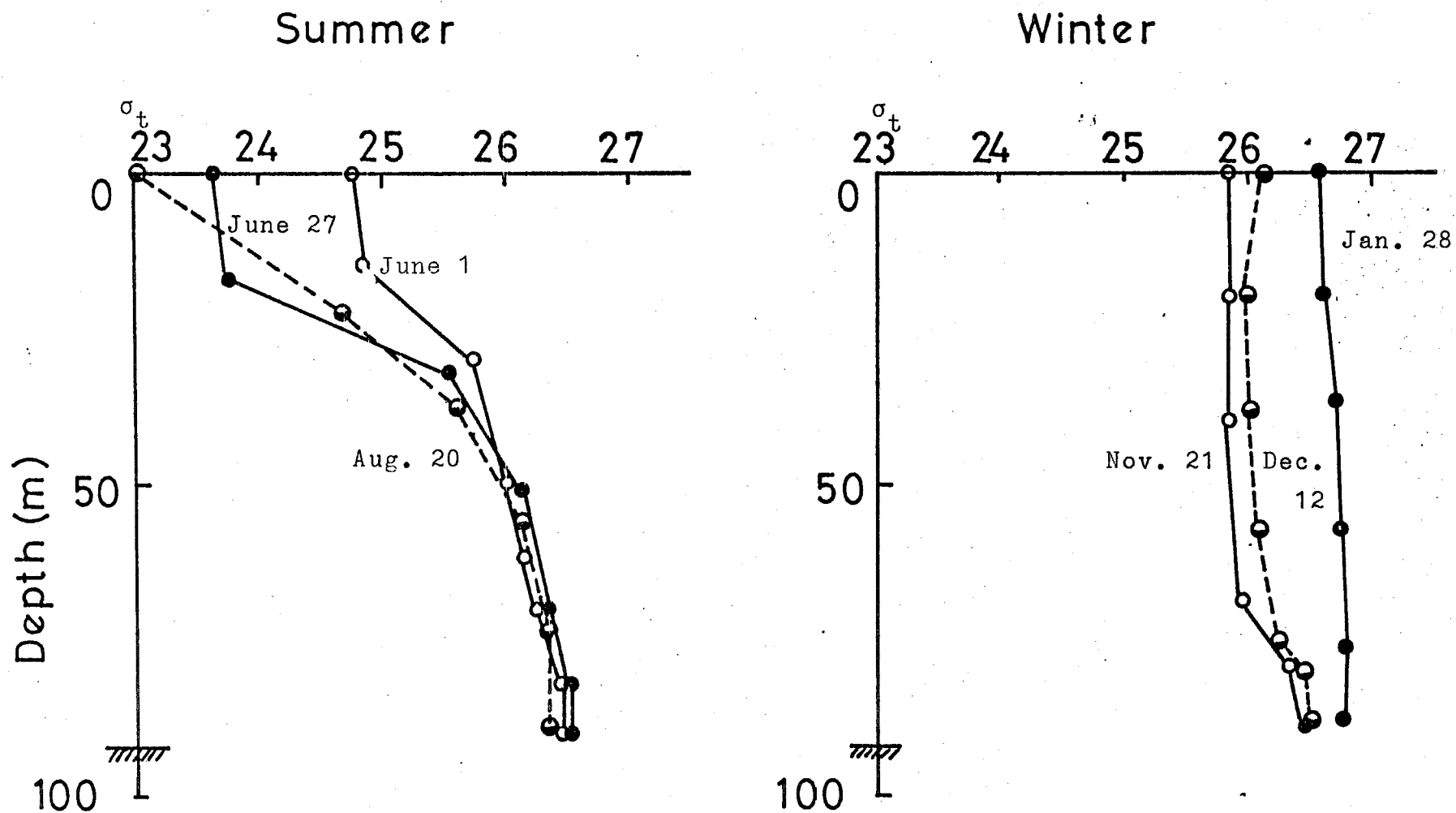


Figure II - 15. Sampling site in Tokyo Bay.

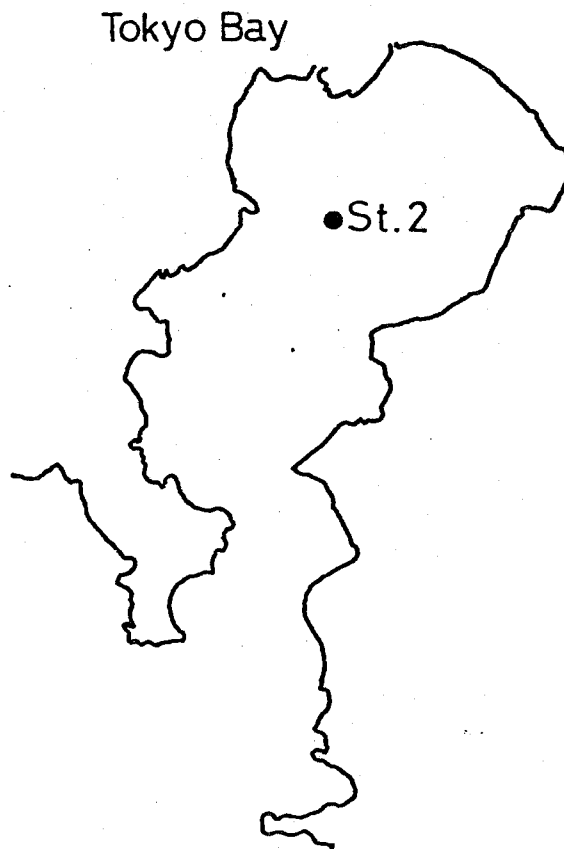
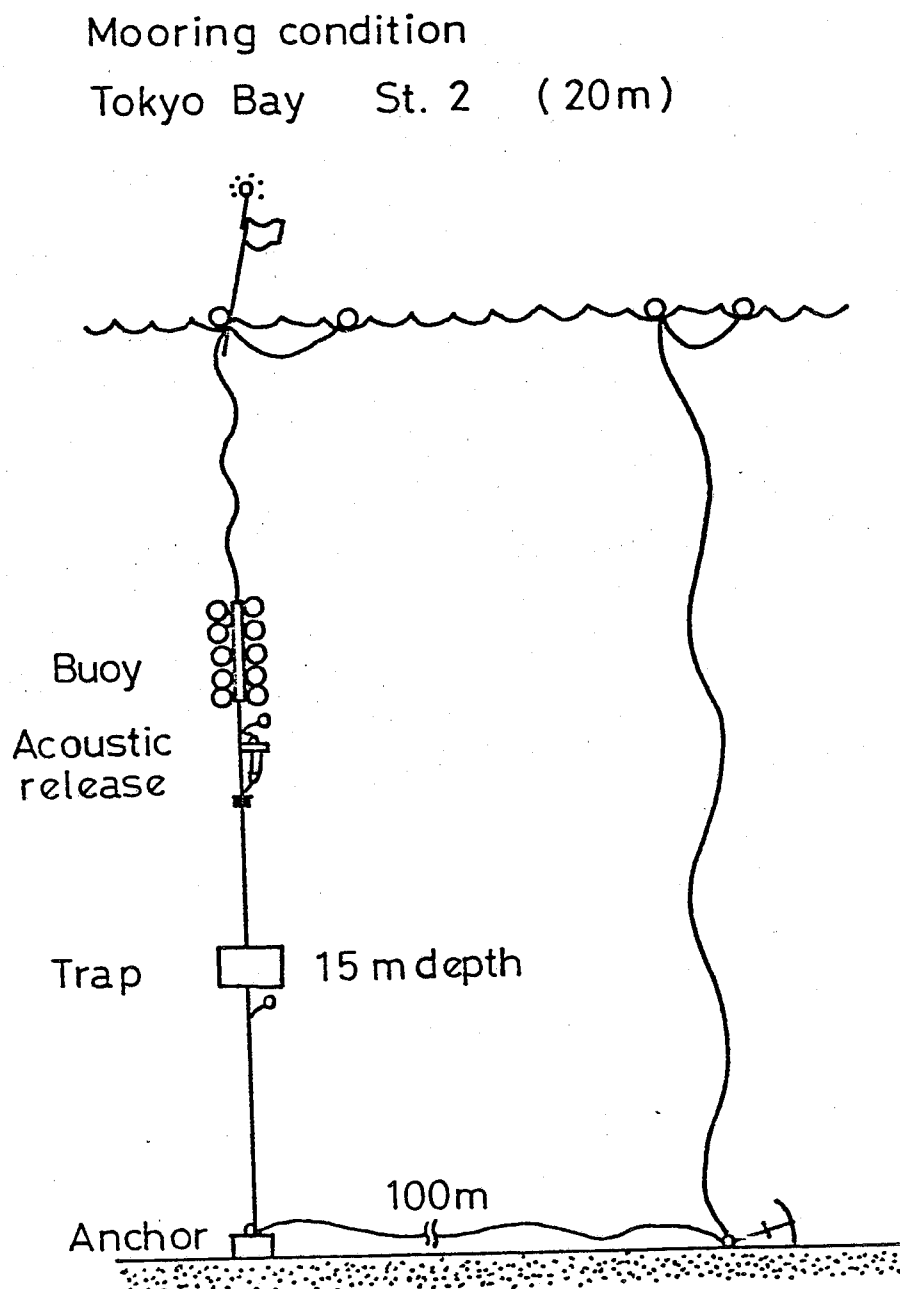


Figure II - 16. Mooring configuration of H(NH) type sediment traps in Tokyo Bay.



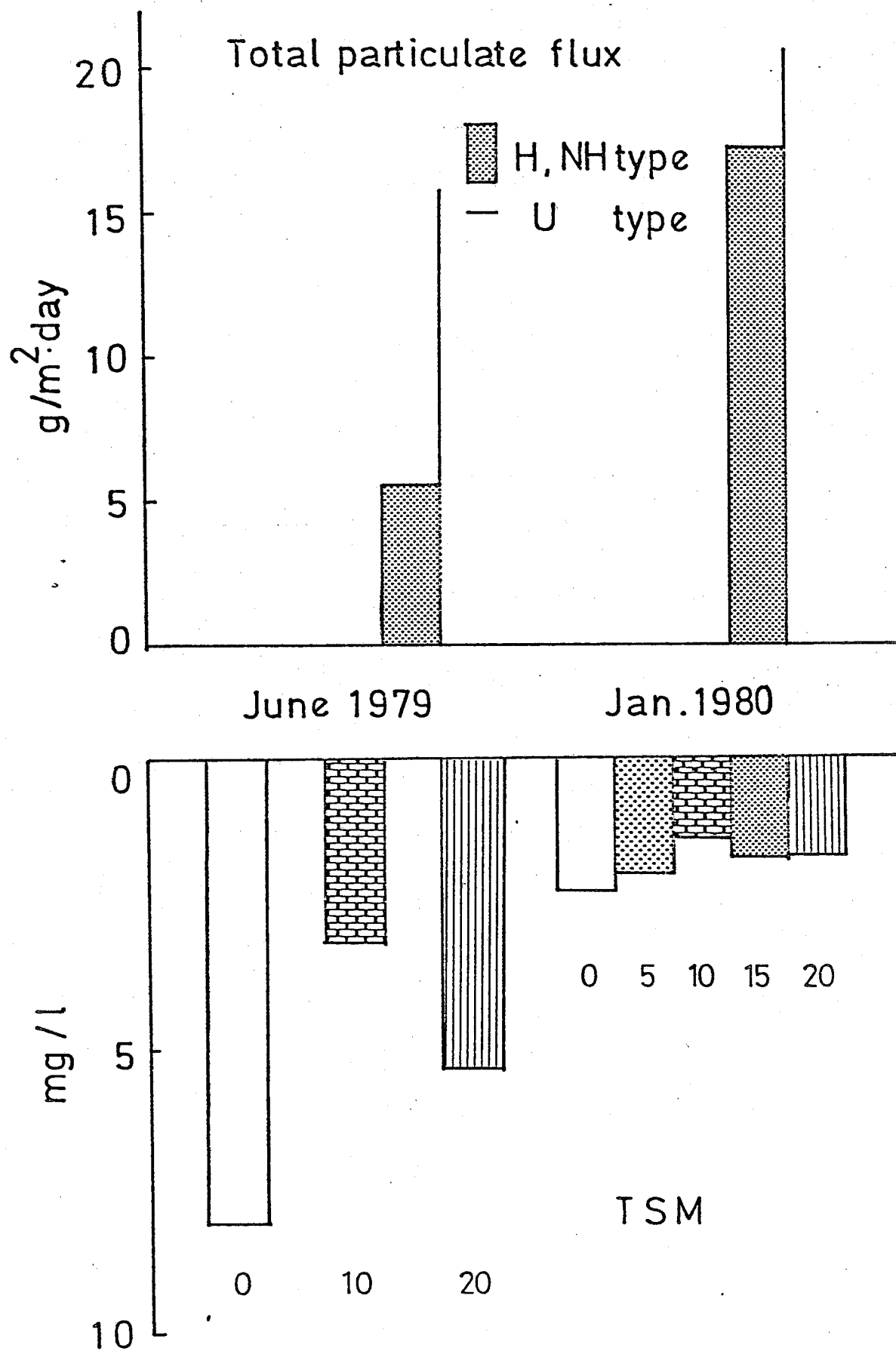


Figure II - 17. Total particulate flux observed with U type and H(NH) type sediment traps and total suspended matter in sea water in Tokyo Bay.

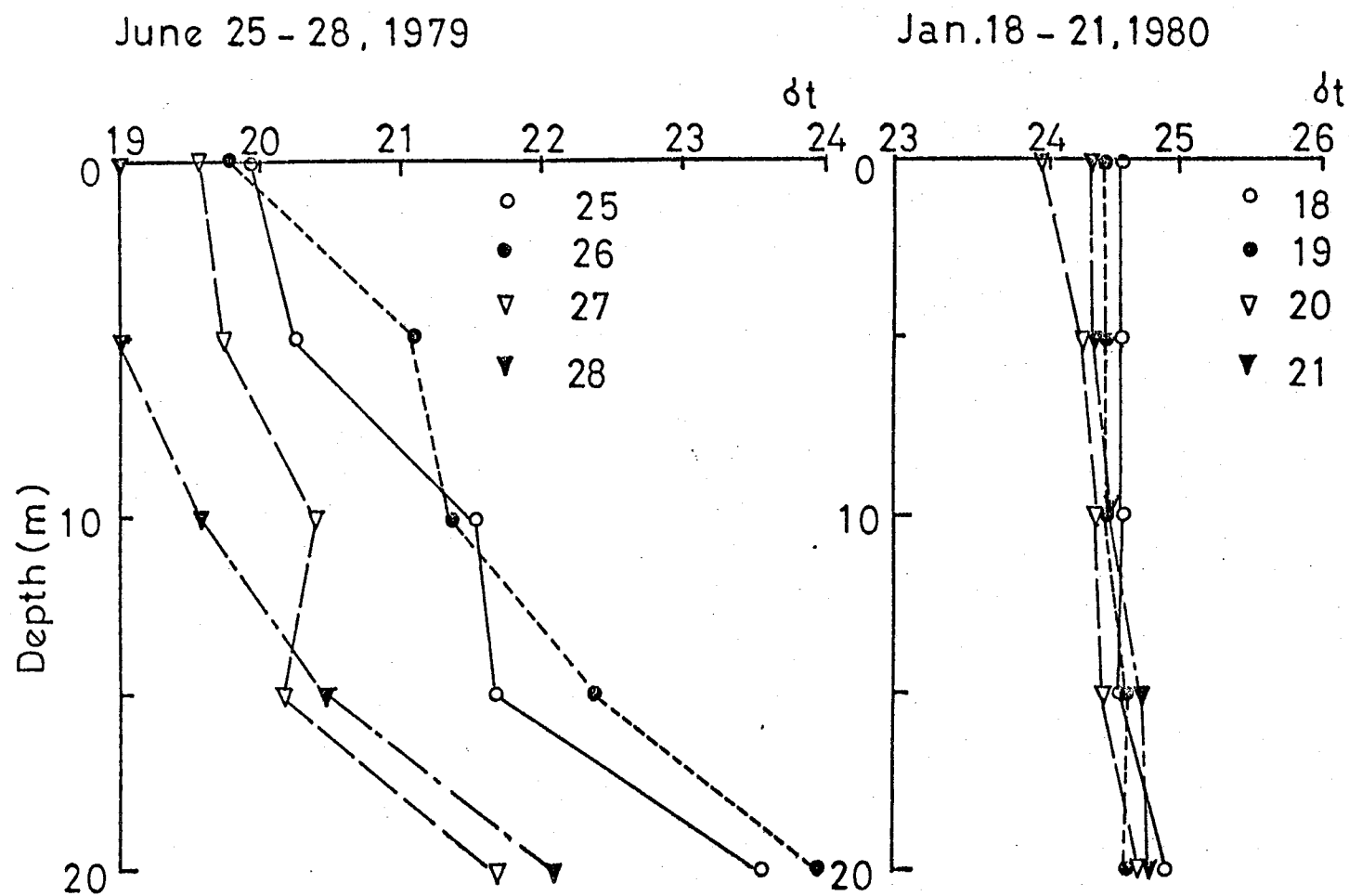


Figure II -18 Vertical profiles of density of water (σ_t) on June, 1979 and Jan., 1980.

Figure II - 19. Vertical profiles of the concentration of Th-234 and chlorinity, specific activity of Th-234 in trapped matter, and the flux of this nuclides.

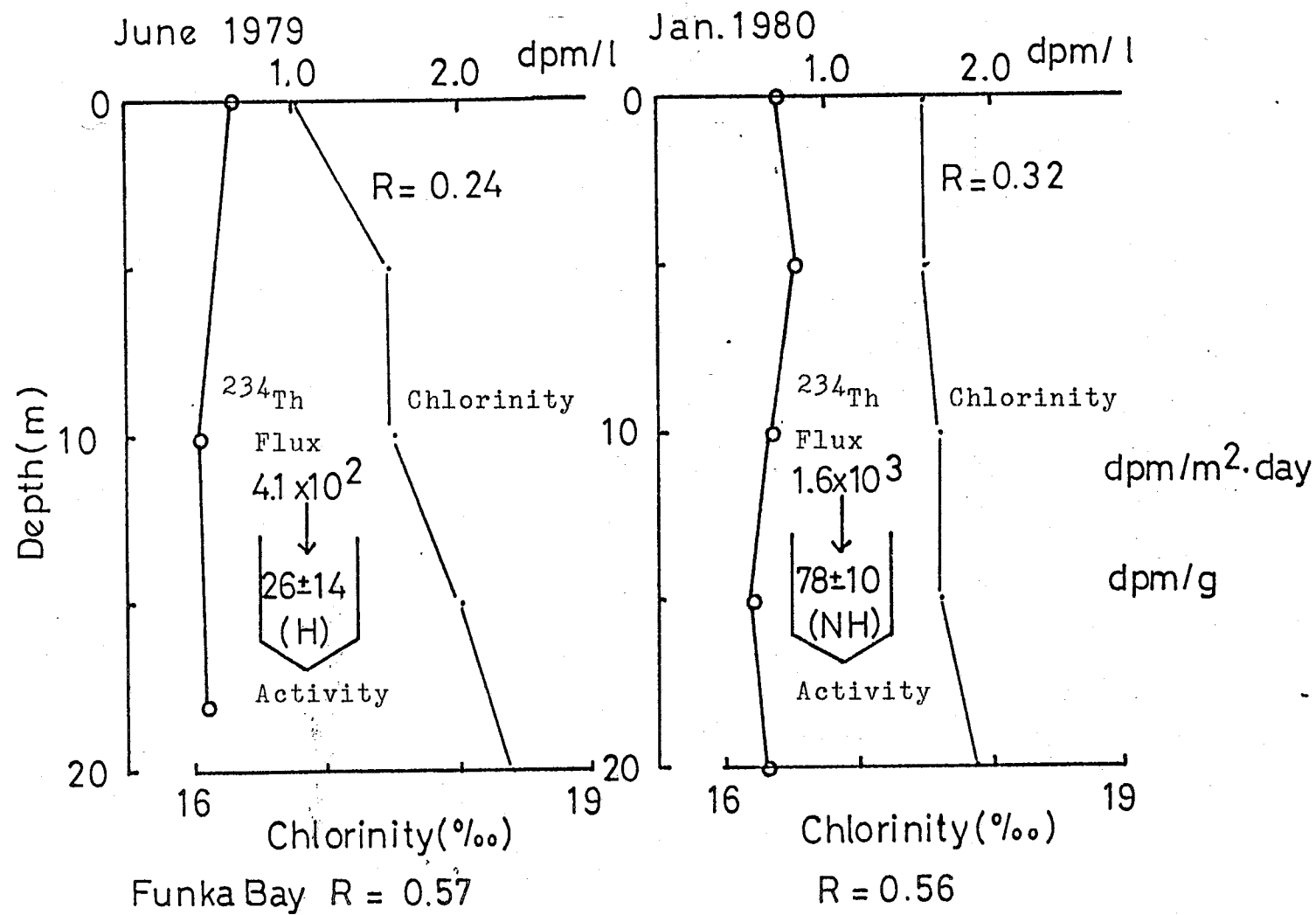


Table II - 1. Concentrations of Be-7, Th-234, Th-228, Th-230 and Th-232 in the settling matter collected with sediment traps at St. P in Funka Bay.

Sampling depth (m)	Specific activity (dpm/gram of dry weight)				
	Be-7 ^{*1}	Th-234 ^{*1}	Th-228	Th-230	Th-232
June 27 - July 23, 1977					
40 m	130	590	1.9±0.1	0.6±0.1	0.4±0.2
80 m	140	1020	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	1770	2.8±0.2	0.5±0.01	1.8±0.2
B	190	2540	0.4±0.1	0.03±0.02	0.1±0.03
80 m F	120	2280	12.6±0.5	1.1±0.2	1.9±0.2
B	220	1320	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 Be-7 and Th-234 are 20 - 30 dpm/g and 40 - 70 dpm/g, respectively.

*2 Cited from Tsunogai et al. (24). 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.

Table II - 2. Activity ratios of Be-7/Th-234, Th-228/Th-234, Th-230/Th-234 and Th-232/Th-234 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 ^{*1}				
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 weighted means of the F and B fractions given in Table 1.

Table II - 3. Observed inventories of Th-234 and Be-7 in sea water and their observed and calculated particulate fluxes at the bottom of each water column.

A. Th-234

Depth (m)	$^{234}\text{Th}/^{238}\text{U}^{*1}$ in sea water(-)	Residence time ^{*1} 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 (22).

B. Be-7

Depth (m)	$^7\text{Be}/^{234}\text{Th}^{*1}$ in settling matter(-)	Residence time ^{*2} 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.

Table II -4. Particulate flux observed at Sta. 2 of Tokyo Bay.
(depth deployed: 20m)

Sample	Total particulate flux ₂ (g/m ² . day)	I.L.* (%)	SiO ₂ * (%)	Al* (%)	Fe* (%)	Mn* (%)	²³⁴ Th (%)
[KT-79-9] Summer							
U type trap	15.85±0.14	31.2	35.8	5.4	4.0	0.21	-
H type trap	5.80±0.41	29.6	39.4	6.3	4.2	0.24	26±14
(sediments)	-	9.0	43.9	5.6	4.7	0.03	-
[KT-80-1] Winter							
U type trap Total	20.71±0.31	19.8	56.3	4.8	3.9	0.27	-
>58μ	7.52	13.9	60.7	6.4	4.7	0.22	-
<58μ	13.19	22.7	53.8	3.9	3.4	0.30	-
NH type trap Total	16.29±1.17	14.0	51.9	4.5	2.9	0.21	78±10
>58μ	5.61	11.3	53.9	6.9	4.6	0.24	-
<58μ	10.68	15.4	50.8	3.3	2.0	0.19	-
(sediments)	-	11.2	36.8	5.2	3.4	0.12	-
[KT-80-16] Summer							
NH type trap Total	1.97±0.64	-	-	-	-	-	-

* After Uematsu (1978)

CHAPTER III

THE VERTICAL TRANSPORT PROCESS IN OPEN SEA

III-1 Introduction

I have proposed in the previous chapter that aggregation and breaking down of particles take place in coastal sea and that fine particles play an important role on the removal process of insoluble heavy metals from sea water. It is necessary to make clear whether this mechanism is important in the open sea or not. I considered it by determining radioisotopes such as Po-210, Pb-210, Th-234 and long lived Th isotopes in sea water and settling matter collected with sediment traps.

III-2 Materials and MethodIII-2-1 Analytical methods

The analytical procedure for Be-7, Th isotopes, Po-210 and Pb-210 in sea water and trapped matter were the same as those used in the chapter II.

III-2-2 Sediment trap employed for this research

H type sediment traps were used at stations in the Northern North Pacific and in Sagami Bay. NH type sediment traps were used at stations in the other sites. The design of the sediment traps are described in the previous chapter.

III-3 Results and Discussion

III-3-1 Be-7 in the ocean water

The concentration of Be-7 in the surface water around Kuroshio was determined during the cruise operated by Hakuho-Maru of University of Tokyo in July, 1977 (KH-77-2). The sampling sites and the results obtained are given in Figure III-1 and Figure III-2, respectively. The vertical distributions of Th-234 in sea water were also determined at three stations (Figure III-3).

The concentration of Be-7 in the surface water near Japan islands is apt to be lower than that far from the land. The activity ratio of Th-234/U-238 obtained is from 0.50 to 0.79, which is also smaller than the activity ratio reported in Northern North Pacific by Matsumoto (1974). I have already shown in the previous section that Be-7 is one of the most insoluble elements which are transported downward by particles and that the residence time of Be-7 is almost the same as that of Th-234 in coastal sea. Assuming that the residence time of Be-7 is equal to that of Th-234 obtained from the disequilibrium with U-238. The input rate of Be-7 (I_{Be}) was estimated with the following equation.

$$I_{Be} = \frac{N_{Be}}{\psi_{Th}}$$

where ψ_{Th} is the residence time of Th-234 in the upper 50 m of water column and N_{Be} is the inventory of Be-7. The results are given in Table III-1. The input rate of Be-7 calculated is 7.4×10^{-2}

- 1.0×10^{-1} atoms/cm²sec. Silker et al. (1974) have estimated the atmospheric input rate of Be-7 to be 3.6×10^{-2} atoms/cm²sec in the Atlantic and Eastern Pacific of the same latitudinal belt. The small difference between them seems to be due to uncertainties of the values used in the both calculations. The input rate of Be-7 reported by Silker et al. (1974) is a mean of widely scattered ones. We can conclude that Be-7 in the open ocean is also influenced by the particulate transport as well as Th-234.

III-3-2 Total particulate flux observed with sediment traps in the open ocean.

Sediment trap experiments in the open ocean were carried out four times from 1978 to 1980. The (NH)H type sediment traps were employed. The mooring configurations of sediment traps are shown in Figure III-4, III-5 and III-6. The sampling sites are given in Figure III-7 and III-8. The observed total particulate fluxes are given in Table III-2. The fluxes observed at all sites increase with depth from the surface to the bottom. This may be thought strange, judging from the following common sense. The particles in sea-water are produced mainly by the biological process in the surface sea-water of the open ocean and reduced gradually by the dissolution and the decomposition during sinking downward to the deep layer. If it is only the process affecting the particulate flux, total particulate flux should decrease with depth monotonously. The above statement includes the following three important hypotheses.

1. The sinking particulate matter is produced only in the surface layer.
2. The effect of horizontal movement of water on the particulate flux is negligibly small.
3. The particulate matter sinks downward unilaterally from the surface to the deep.

These conditions can not be accepted easily as facts. I am going to show that these are not valid in the later sections.

The sediment trap experiments at the site 60 miles off the east of Shiriya Zaki were carried out twice on June 2, 1978 and on May 11, 1980. The particulate fluxes observed showed extremely different values. The traps deployed on May 11, 1980 probably caught the particles produced by the blooming of phytoplankton in spring. The total suspended matter observed on May 11, 1980 also was twofold higher than that observed on June 2, 1978 even at 1000 m depth. (Figure III - 10)

Honjo et al. (1980) have energetically carried out the sediment trap experiments in the open ocean. They have used funnel type sediment traps with honeycombs. The total particulate fluxes reported by them were from 0.013 to 0.069 $\text{g/m}^2\text{day}$ and the fluxes observed in this research at northern North Pacific were 0.034 - 0.21 $\text{g/m}^2\text{day}$. The difference between the two might be caused by the effect of the shape of traps employed as discussed by Gardner (1979). The H(NH) type traps used in this research are of a cylinder type shown in Figure II -6 and Figure II -7.

III-3-3 Radioactive nuclides in the sediment trap samples in the open sea.

The specific activities of Be-7, Th-234, Po-210, Pb-210, Th-228, Th-230 and Th-232 in settling material collected with sediment traps are given in Table III -3. The specific activities of all nuclides observed off the Shiriya Zaki on May 11, 1980 increased with depth monotonously. However, the specific activities of these nuclides observed at Sagami Bay were almost constant or decreased with depth, except those of Po-210. The nuclides determined in northern North Pacific can be classified into two types. One is the type that the specific activity increases with depth monotonously and the other first decreases from the surface to the deeper water and increases to the bottom. Po-210 and b-210 belong to the former and the latter includes long-lived Th isotopes. This tendency was not always seen in the results obtained in tropical Atlantic by Brewer et al (1980). All radioisotopes measured by them show the tendency increasing with depth. The difference between them may not be explained only by the spacial variability. It is known that the shape of a trap employed largely affects the amount of material collected as discussed by Gardner (1980). Therefore I suggest that the difference in the trap geometry affects also the composition of material collected.

Tsunogai et al. (1980) made a funnel type trap (L type) which was almost the same shape as used by Honjo (1980). The matter collected with the trap in Funka Bay in summer consisted only of black and dense large particles and its amount was one

order of magnitude smaller than that of observed by the U type trap. This may be due to a fact that the L type trap did not catch so much the fine particles as the U type trap did. If, therefore, the chemical composition is a function of the particle size, the composition will vary with the shape of a sediment trap used. However, We do not know precisely the degree of the variation in the trapping efficiency in the deep pelagic ocean.

III -3-4 Net removal flux and observed flux with sediment traps.

The fluxes of radioisotopes observed with sediment traps in this research are given in Table III -4. In this section, the observed flux with sediment traps will be compared with the net removal fluxes calculated from the disequilibria with their respective parent nuclides. The same method mentioned in the chapter II.

The mass balance of such an insoluble nuclides in the layer of d m thickness is expressed as follows,

$$d \frac{dN_d}{dt} = I - d \cdot \lambda_d \cdot N_d - d \cdot k \cdot N_d \text{ ----- (1)}$$

where N_d is the concentration of a daughter radionuclides in sea water, I is input rate of the nuclides from the atmosphere and/or production rate of it from its parent nuclide in sea water, λ is the radioactive decay constant and k is the removal rate constant. The assumption of a steady state reduces the left term to be 0.

The downward flux (net removal flux), F_d , across the d m of water depth is expressed as:

$$F_d = d \cdot k \cdot N_d \text{ ----- (2)}$$

From the equation (1), we get:

$$N_d = \frac{I}{\lambda_d + k} \text{ ----- (3)}$$

Substituting this equation to the equation (2), we obtain:

$$F_d = d k \frac{I}{\lambda_d + k} \text{ ----- (4)}$$

The observation of the accurate concentration of Th-230 in sea water is very limited because of its extremely low concentration. However, the distribution of U-234, which is a parent of Th-230, is uniform in sea-water and, above all, the long half-life of Th-230 (7.5×10^{10} years) makes its removal flux to be equal to its production rate in the water column. The equation (4) for Th-230 can be simplified as follows:

$$I = d \cdot \lambda_p \cdot N_p, \quad k = 0.01 - 0.001 \text{ yr}^{-1} \gg \lambda_d = 8.6 \times 10^{-12} \text{ yr}^{-1}$$

$$\frac{k}{\lambda_d + k} = 1$$

$$F_d = d \cdot \lambda_p \cdot N_p \frac{k}{\lambda_d + k} = d \cdot \lambda_p \cdot N_p \text{ ----- (5)}$$

The equation (5) is very useful, because the flux is a function of only its concentration regard less of its k value. The concentrations of Ra-226 and Pb-210 near the trap site have been reported by Nozaki and Tsunogai (1976). They have estimated the residence time of Pb-210 to be about 100 years. Considering the atmospheric input of Pb-210 to the surface ocean, the equation (4) is rearranged:

$$F_d = d \cdot \lambda_d \cdot (1/R - 1) \cdot N_d + I \text{ ----- (6)}$$

where I is the input rate of Pb-210 from the atmosphere and R is the activity ratio of Pb-210/Ra-226. Assuming the atmospheric input rate of Pb-210 to be 1.8 dpm/cm²year (Shinagawa, 1980), We have calculated the net removal flux of this nuclide. The result of the calculation for Pb-210 is plotted with the observed flux with sediment traps vs depth in Figure III -11 and that of Th-230 is shown in Figure III -12.

The fluxes of both nuclides exceed the respective net removal fluxes in the upper layer and are almost the same as or a little smaller than that the net fluxes calculated in the deeper layer. This means that in the upper layer the surpluses of Th-230 and Pb-210 are transformed to the particles which can sink downward.

Why the surplus fluxes of these nuclides are observed with sediment traps? It is easy to be explained by the Tsunogai and Minagawa's model(1978). This model suggests that insoluble these nuclides ^{can be} chiefly transported downward by the particles which easily grow up to the large aggregates and break down to small particles. The large aggregates gravitationally sink downward with a significant speed, but the particles broken to small sizes have negligibly small settling velocity and then move up with an eddy diffusion and upwelling of water. Lal and Larman (1973) suggested that the particles smaller than 0.45 microns in diameter move upward under the velocity of upwelling water of 1.4×10^{-5} cm/sec by using Stokes' law. The sediment traps seem to collect the larger particles more effectively and often give different fluxes from the net removal fluxes of these nuclides and insoluble heavy metals.

The fluxes observed at Sagami Bay and a site off the Shiriya Zaki also exceed the net removal fluxes estimated from the concentrations of these nuclides in sea water. The residence time of Th-234 in the surface water of the open sea is around 100 days (Matsumoto, 1975). This value means that the net removal flux of Th-234 from the water column of 320 m thick is ca. 2.0×10^3 dpm/m²day. The net fluxes are calculated to be 0.08 dpm/m²day for Th-230 and 60 dpm/m² day for Pb-210 across the 1200 m surface. All the observed fluxes, except those of Th-234 and Pb-210 observed at the 320 m surface in Sagami Bay, exceed the net fluxes given above. (Table III -4)

Brewer et al. (1980) have also observed the surplus fluxes of Th-230 and Pa-231 obtained with the sediment traps

in Panama Basin. The sediment traps designed by Honjo (1978) was employed in the experiment. They suggested that this results was of a special case and have explained that the surplus fluxes of Th-230 and Pa-231 were due to the horizontal transport of these nuclides from the high productive area (upwelling region). However, the apparently over-trapping of these nuclides seems to be a common phenomenon in the oceans as the same fact has been found also in the Pacific. This fact is also an evidence showing the occurrence of the considerable "upward flux" of these nuclides in the oceans.

Table II - 5. Net removal flux and observed flux in Funka Bay and Tokyo Bay.

		Summer	Winter
Tokyo Bay (20m)	R	0.238	0.318
	τ (days)	11	16
	F (dpm/m ² ·day)	7.1×10^2	6.6×10^2
	F:trap	4.1×10^2	1.6×10^3
Funka Bay (90m)	R	0.570	0.559
	τ	46	45
	F	4.4×10^2	4.4×10^2
* Narragansett Bay(10m)	R	0.174	0.425
	τ	7	26
	F	—	—

* P.H. Santshi (1979)

Table III - 1 Mean concentrations of Th-234 and Be-7 in the surface water of Kuroshio and the input rate of Be-7 estimated using a residence time of Th-234.

Sampling station	Th-234 (dpm/l)	Residence time (day)	Be-7 (dpm/1000l)	Estimated input rate of Be-7 (atoms/cm ² sec)
St. 16	2.0	175	870	1.0×10^{-1}
St. 41	1.4	45	470	1.1×10^{-1}
St. 78(14)	1.2	35	280	7.4×10^{-2}
				$(3.6 \times 10^{-2})^*$

* W. B. Silker (1974)

Table III - 2 Total particulate fluxes observed with sediment traps in Sagami Bay
and open sea ($\text{g/m}^2\text{day}$).

Shiriyu Zaki (KT-78-8)		North Pacific (KH-78-3)		Sagami Bay (KT-79-12)		Shiriyu Zaki (KT-80-6)	
Depth (km)	Flux	Depth (km)	Flux	Depth (km)	Flux	Depth (km)	Flux
0.32	0.177	0.1	0.055	0.32	0.69	0.15	3.02
1.2	0.277	1.1	0.208	1.2	2.87	1.06	5.53
		2.2	0.196			1.29	4.97
		4.4	0.148				
		5.25	0.034				
Anchor Depth	1260 m		5300 m		1430 m		1340 m

Table III - 3 Specific activities of Th-234, Be-7, Po-210, Pb-210, Th-228, Th-230
and Th-232 in ^{the} settling material collected with sediment traps.

Depth (m)	Th-234*	Be-7*	Po-210*	Pb-210*	Th-228*	Th-230*	Th-232*
Sagami Bay (KT-79-12) July 24, 1979							
320	2350	-	85	85	4.5	0.4	0.4
1200	2100	-	120	61	2.3	0.5	0.4
Shiriyu Zaki (KT-80-6) May 11, 1980							
150	850	-	10	27	3.4	0.3	0.2
1060	-	-	20	27	-	-	-
1285	2230	-	26	58	6.4	0.4	0.3
North Pacific (KH-78-3) Aug. 14, 1978							
100	-	<286@	15	14	7.2	1.8	1.2
1100	-	< 57@	93	66	3.5	0.7	0.5
2200	-	<141@	193	123	2.4	0.4	0.2
4400	-	<100@	650	144	2.6	0.9	0.4
5250	-	<400@	921	421	5.6	7.0	0.7

* dpm/g , @ these are upper limit values estimated from the detection limit.

Table III - 4 Fluxes of Be-7, Th-234, Po-210, Pb-210, Th-228, Th-230 and Th-232 observed with sediment traps.

Depth	Th-234	Be-7	Po-210	Pb-210	Th-228	Th-230	Th-232
(m)	(dpm/m ² day)						
Sagami Bay							
320	1.6x10 ³	-	58	58	3.1	0.3	0.3
1200	5.1x10 ³	-	83	175	6.6	1.2	1.1
Shiriya Zaki							
150	2.6x10 ³	-	31	82	10.3	0.8	0.5
1060	-	-	110	150	-	-	-
1285	1.1x10 ⁴	-	130	288	32.0	2.2	1.3
North Pacific							
100	-	-	0.8	0.8	0.4	0.1	0.1
1100	-	-	19.3	14.0	0.7	0.1 ₄	0.1
2200	-	-	38	24	0.5	0.1	0.04
4400	-	-	96	21	0.4	0.1 ₄	0.06
5250	-	-	31	16	0.2	0.3	0.02

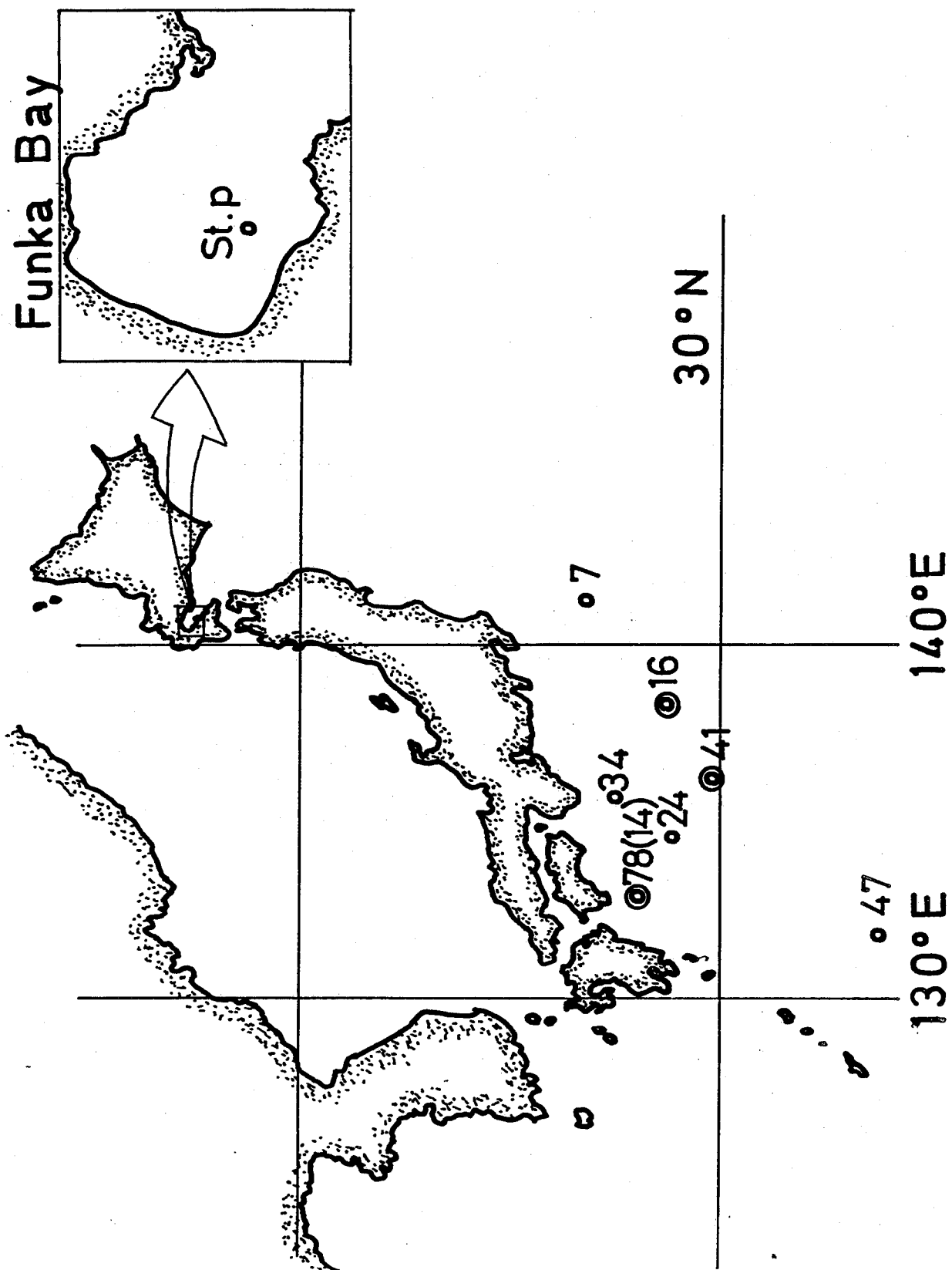


Figure III - 1. Sampling locations

Figure III - 2. Concentrations of Be-7 in the surface water.

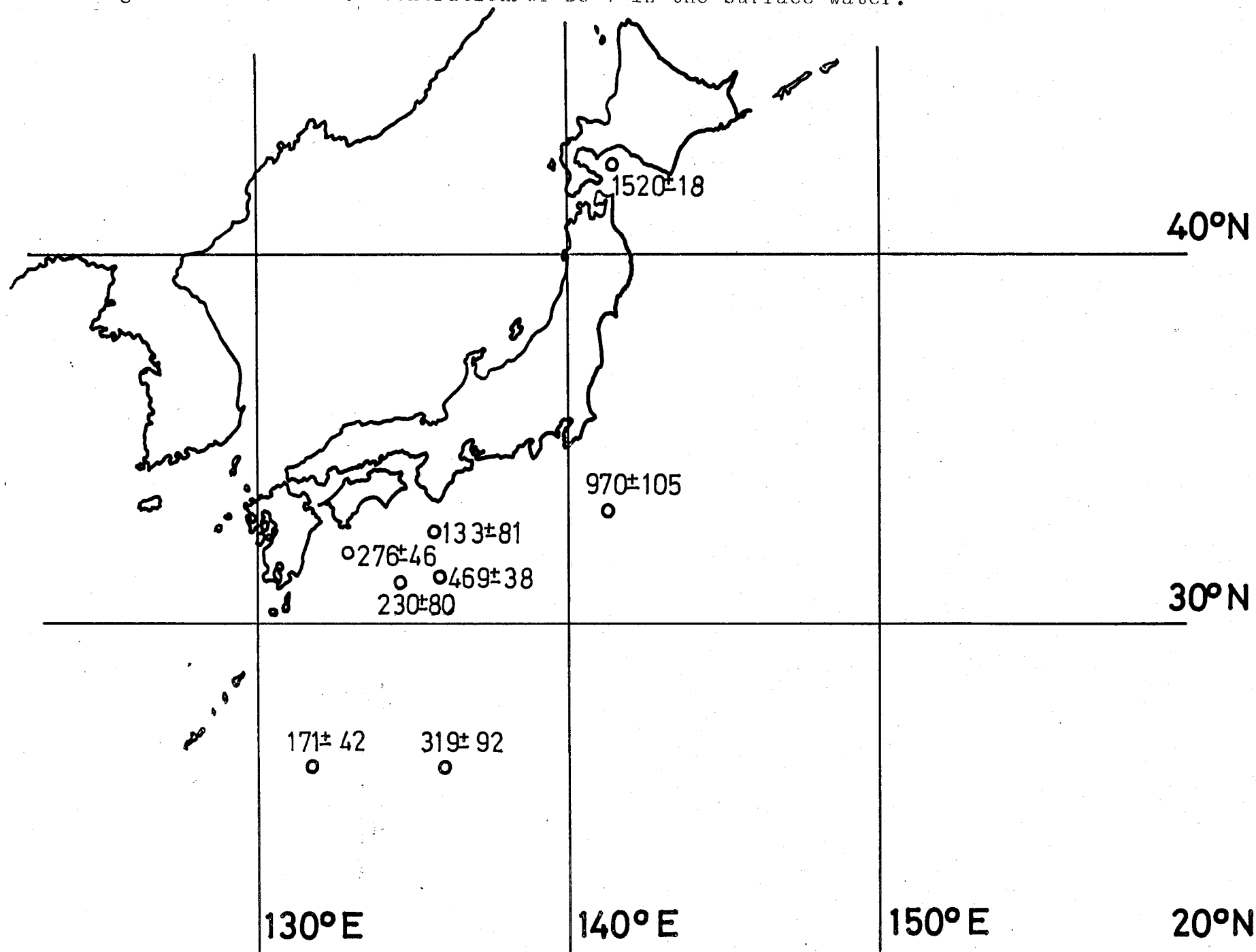


Figure III - 3. Vertical profiles of Th-234 in the surface water.

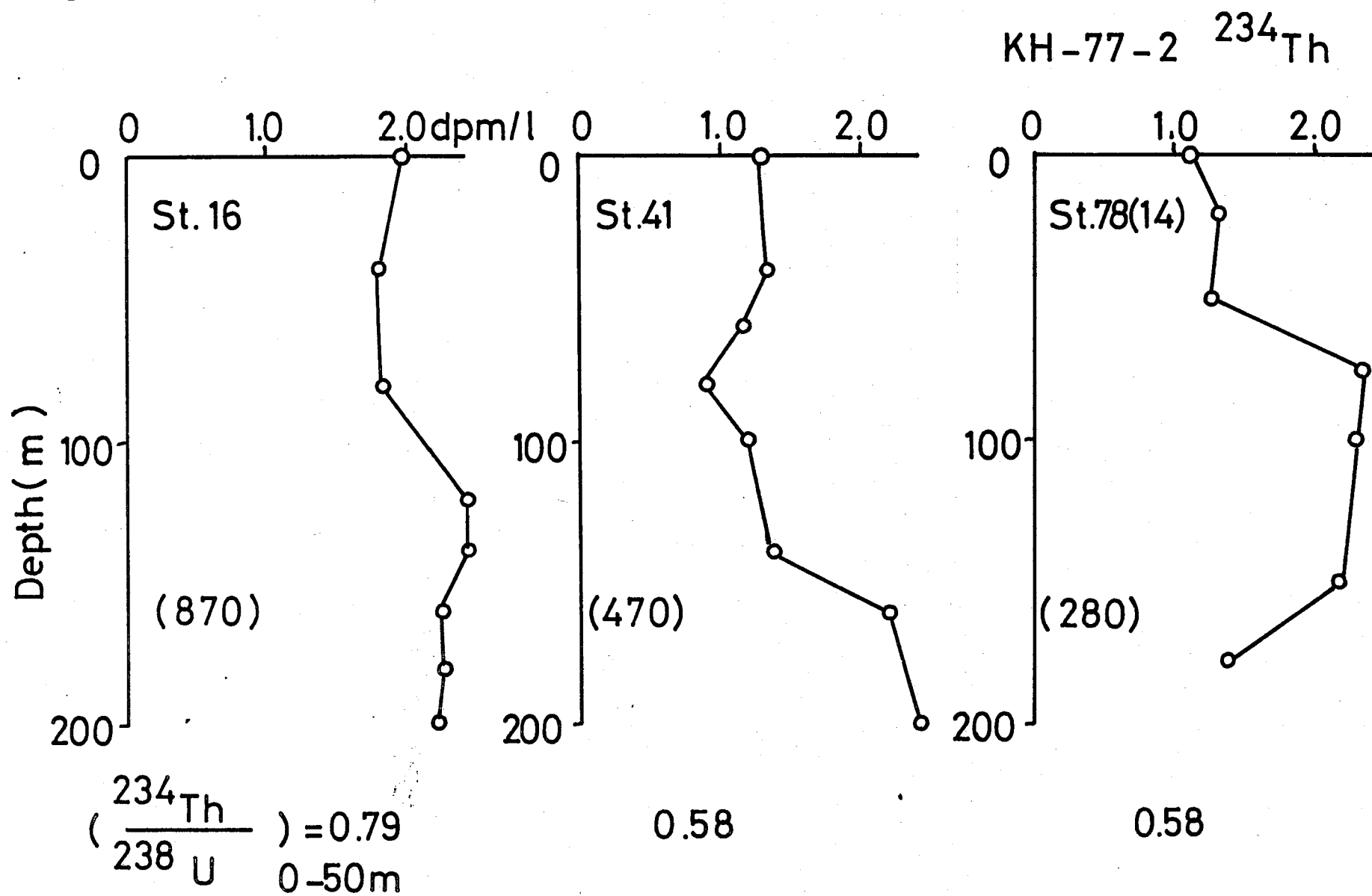


Figure III - 4. Mooring configuration of sediment traps at Sagami Bay in 1979.

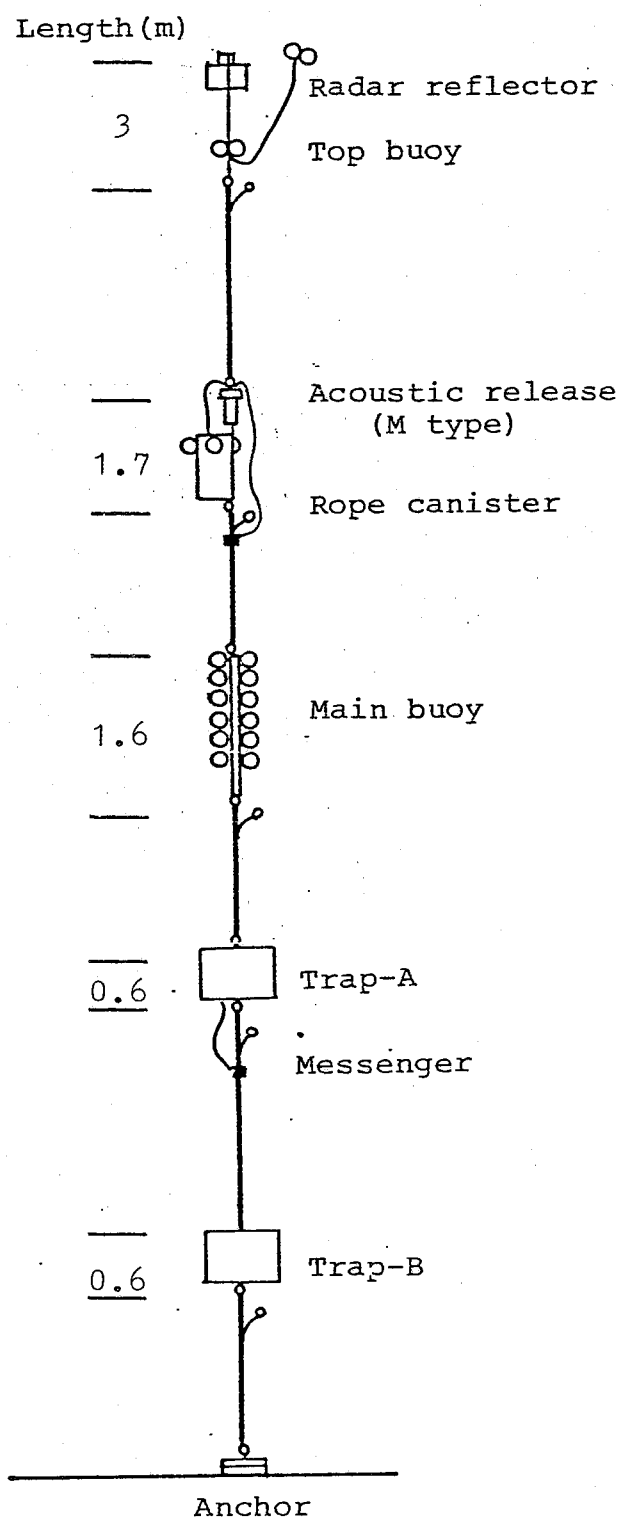
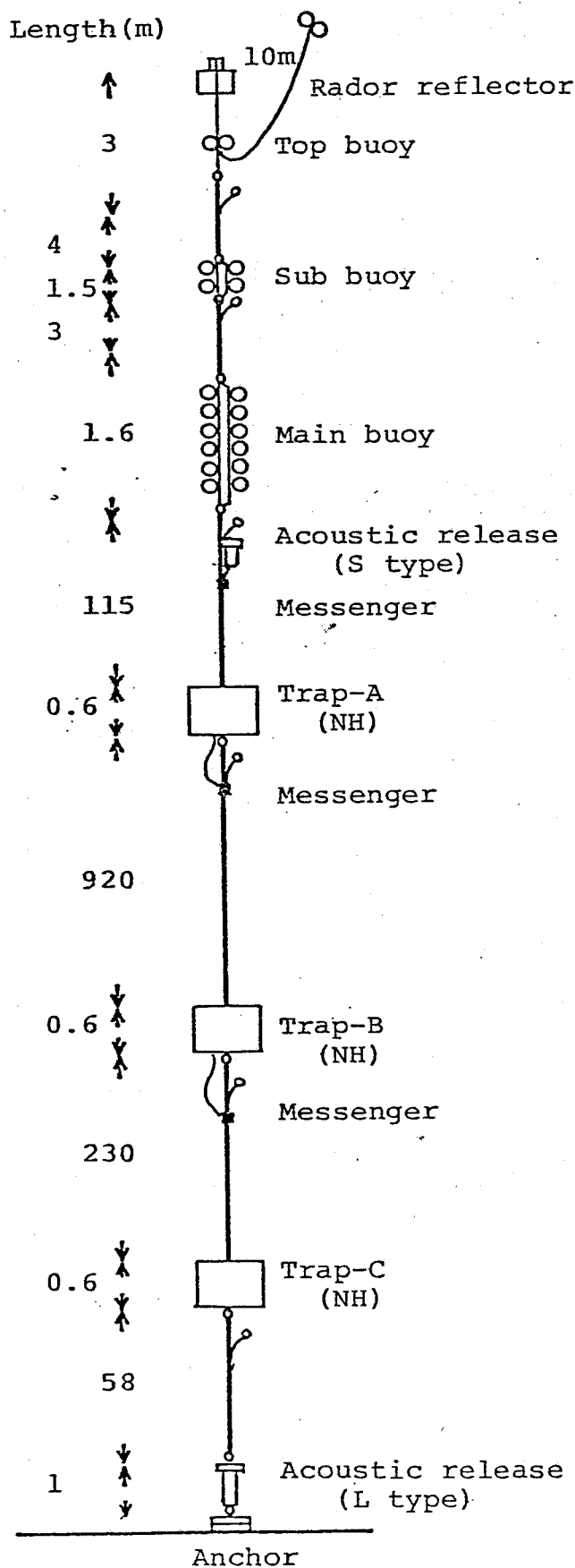


Figure III -5. Mooring configuration of sediment traps at the site
60 miles off the east of Shiriya Zaki.

-80-

KT-80-6 May 11 - May 14, 1980

SYSTEM 1



SYSTEM 2

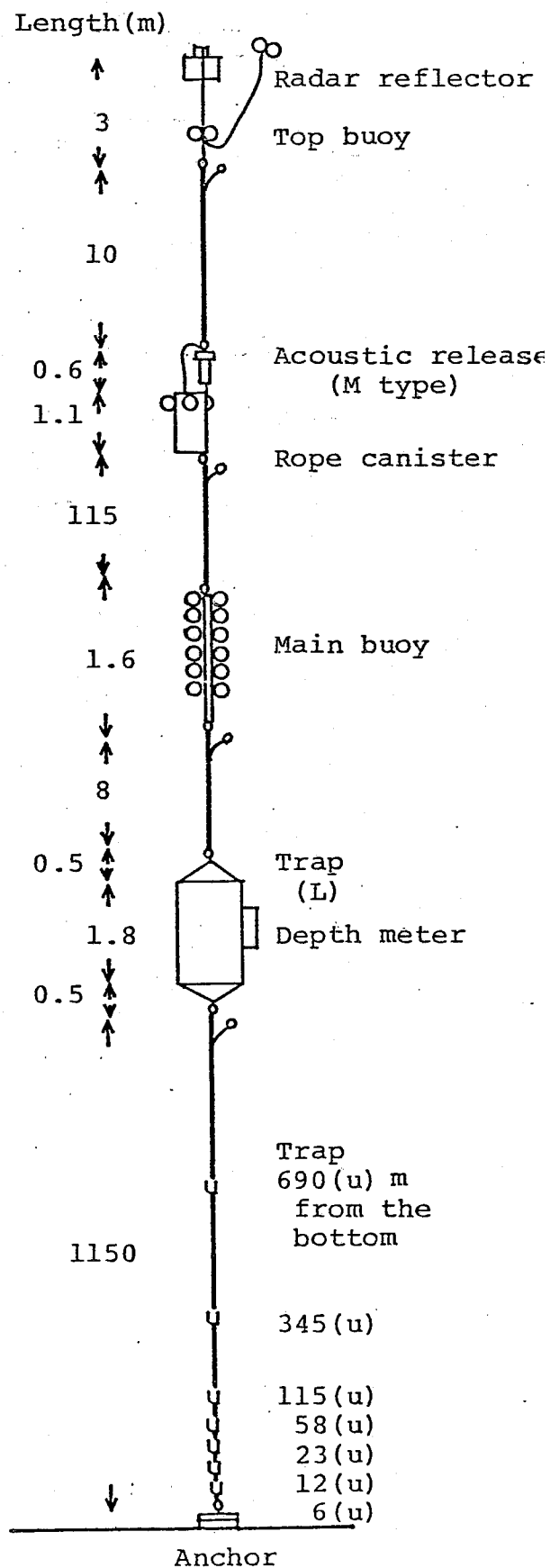
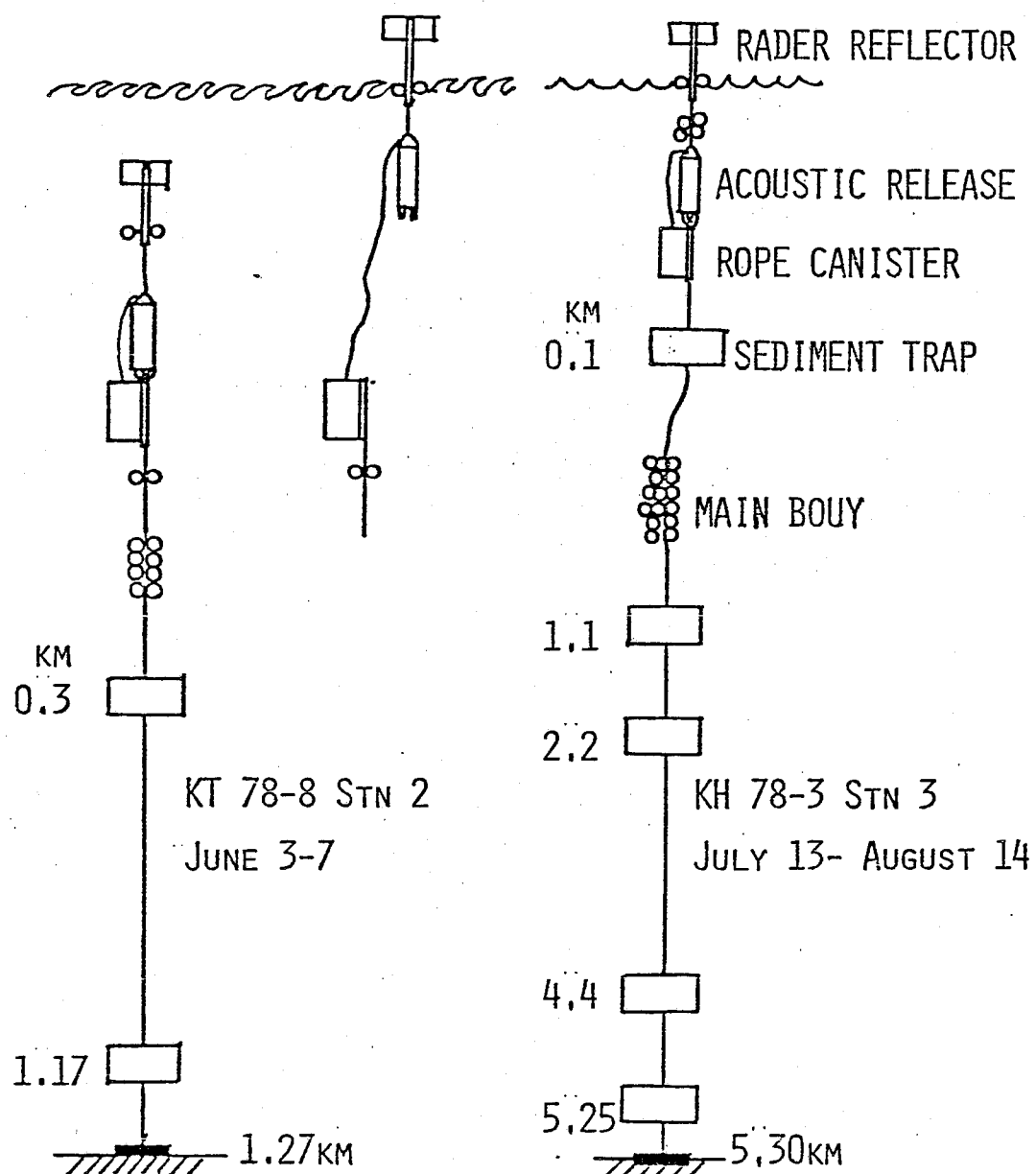


Figure III - 6. Mooring configuration of sediment traps at the site 60 miles off the east of Shiriya Zaki in 1978 and in northern North Pacific.



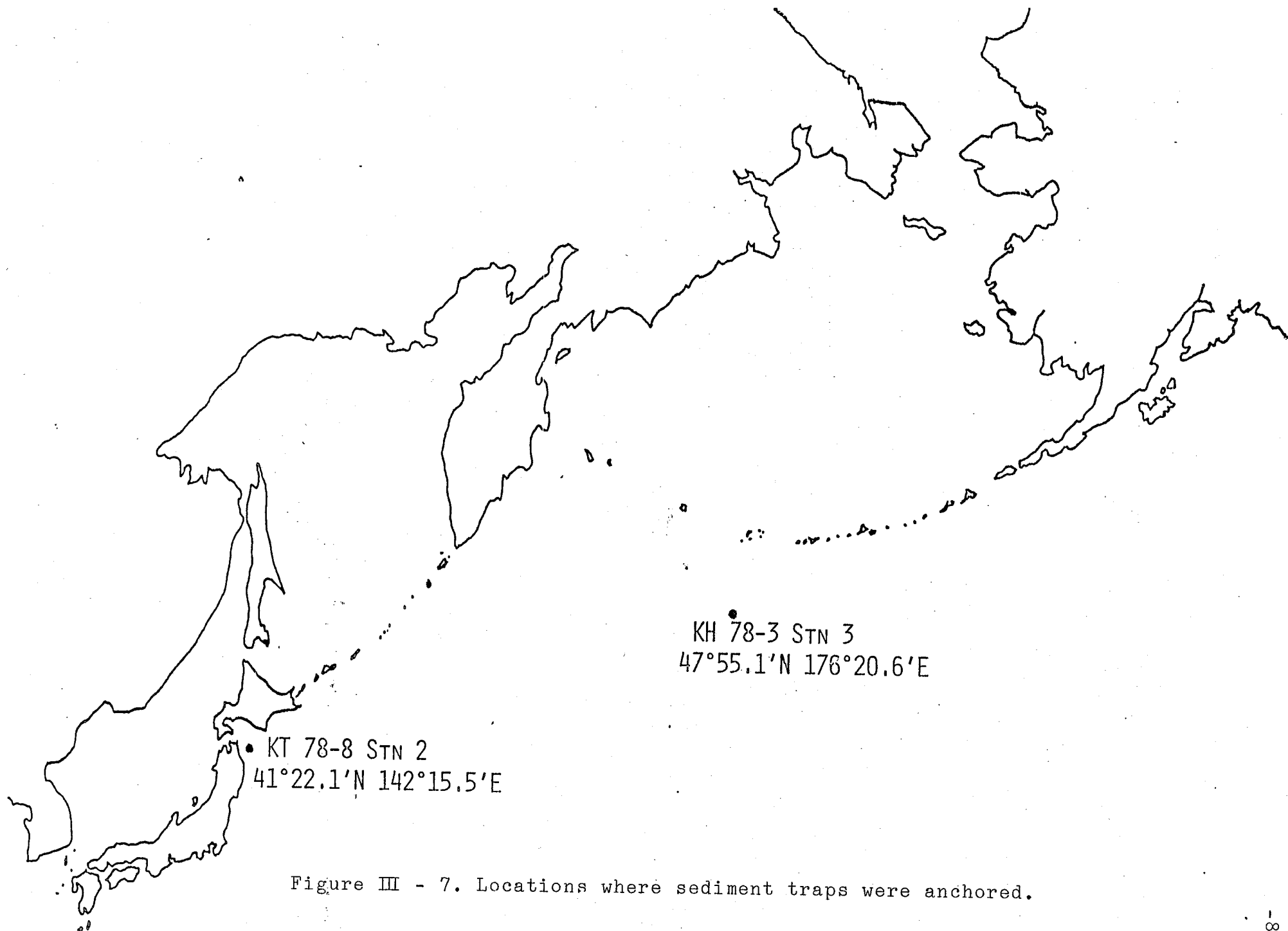


Figure III - 7. Locations where sediment traps were anchored.

Figure III - 8. Sampling site in Sagami Bay.

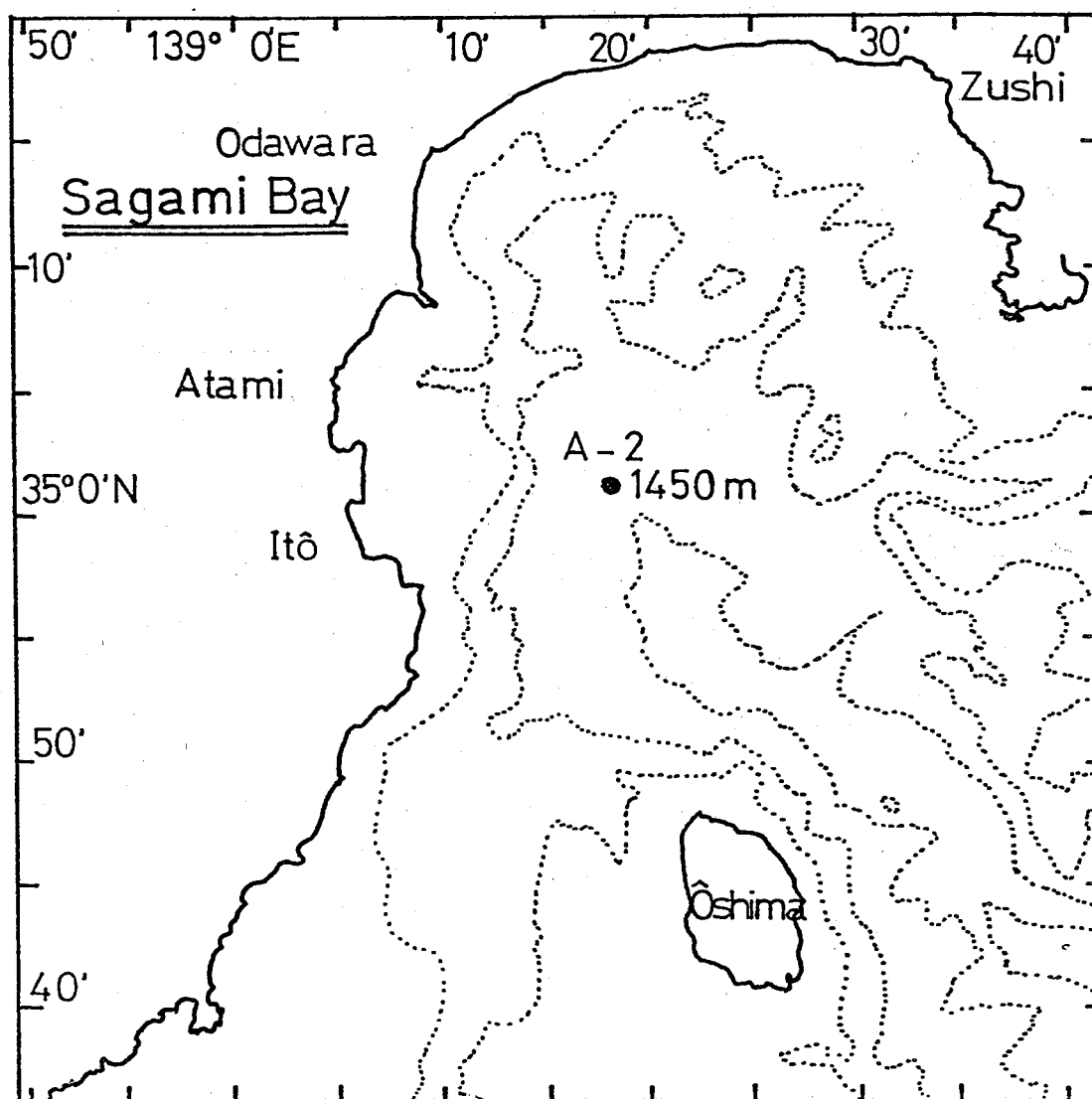


Figure III - 9. Temperature, salinity and total suspended matter at St. A-2 in Sagami Bay on July 28, 1979.

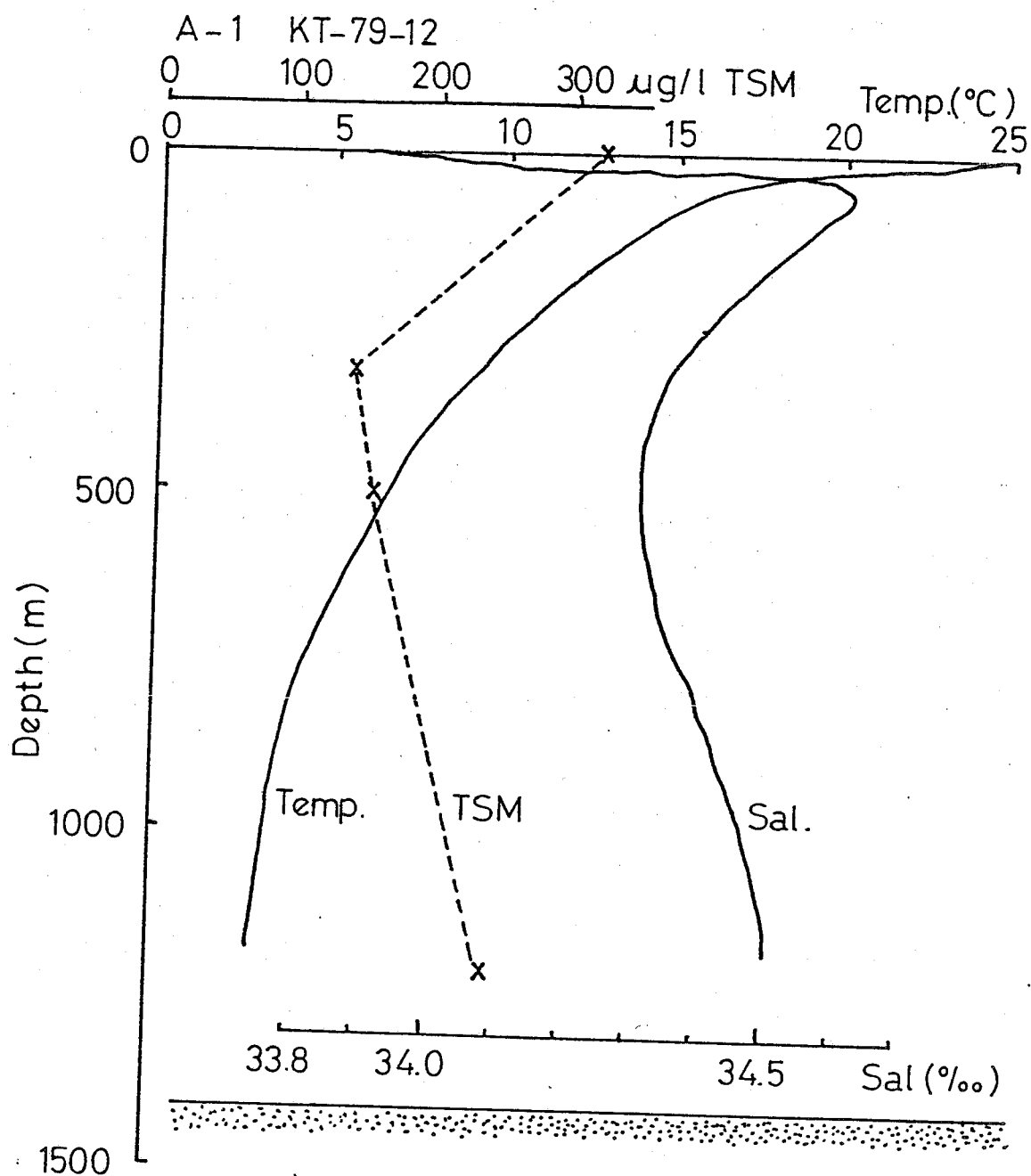


Figure III - 10. Total suspended matter at the site
60 miles off the east of Shiriya Zaki
on June 2, 1978 and May 11, 1980.

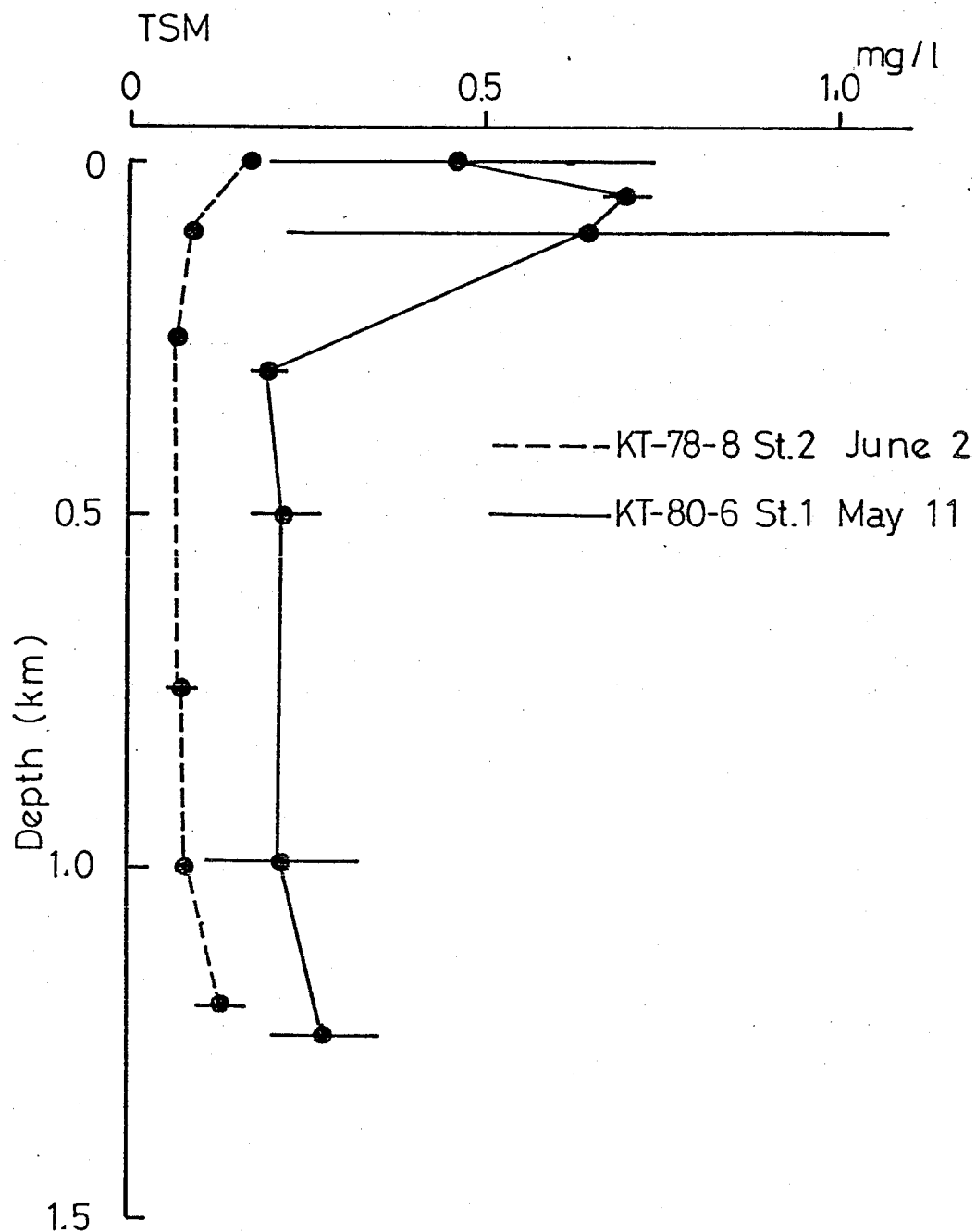


Figure III - 11 Flux of Pb-210 observed with sediment traps in North Pacific and its net removal flux calculated.

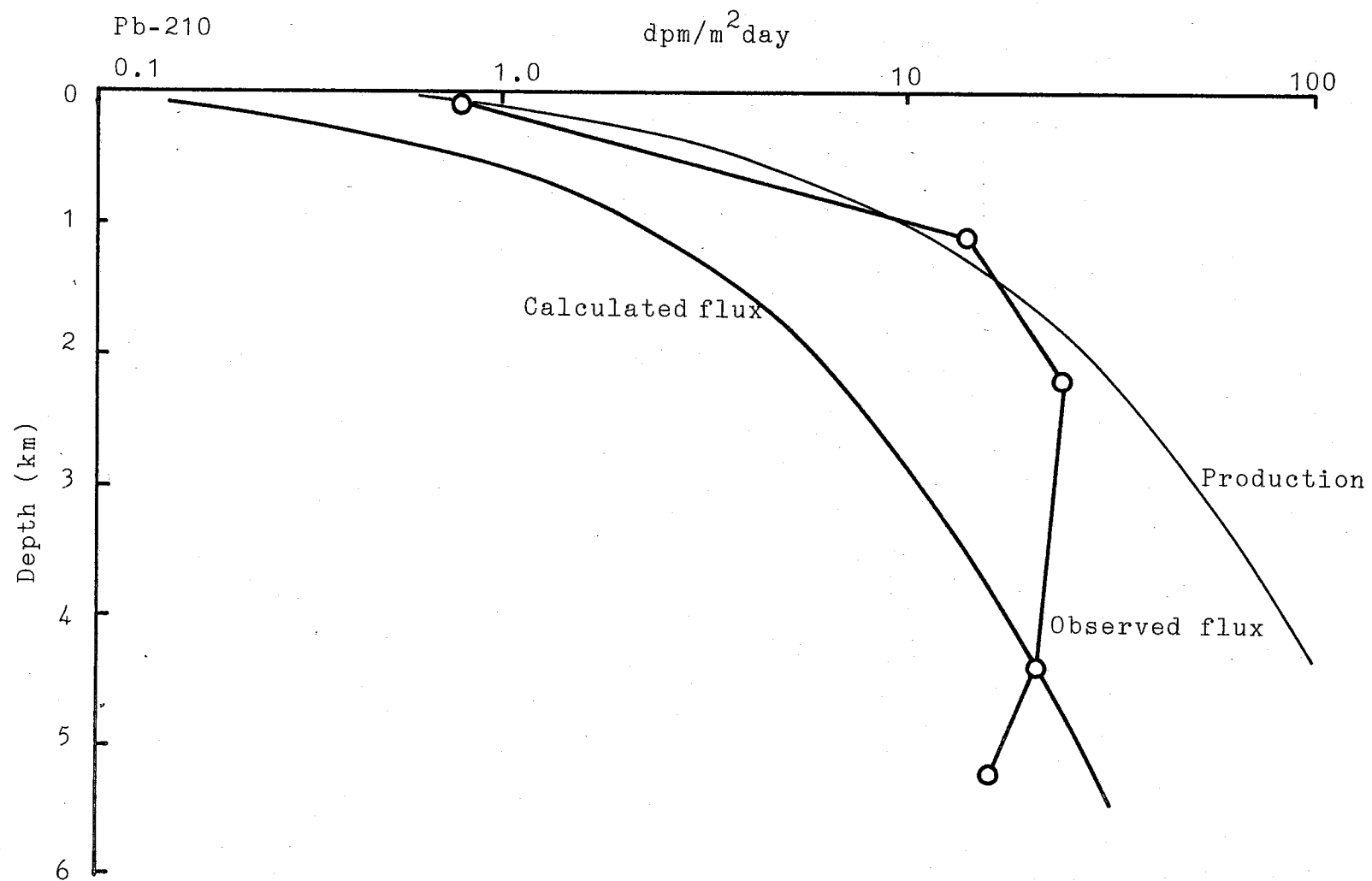
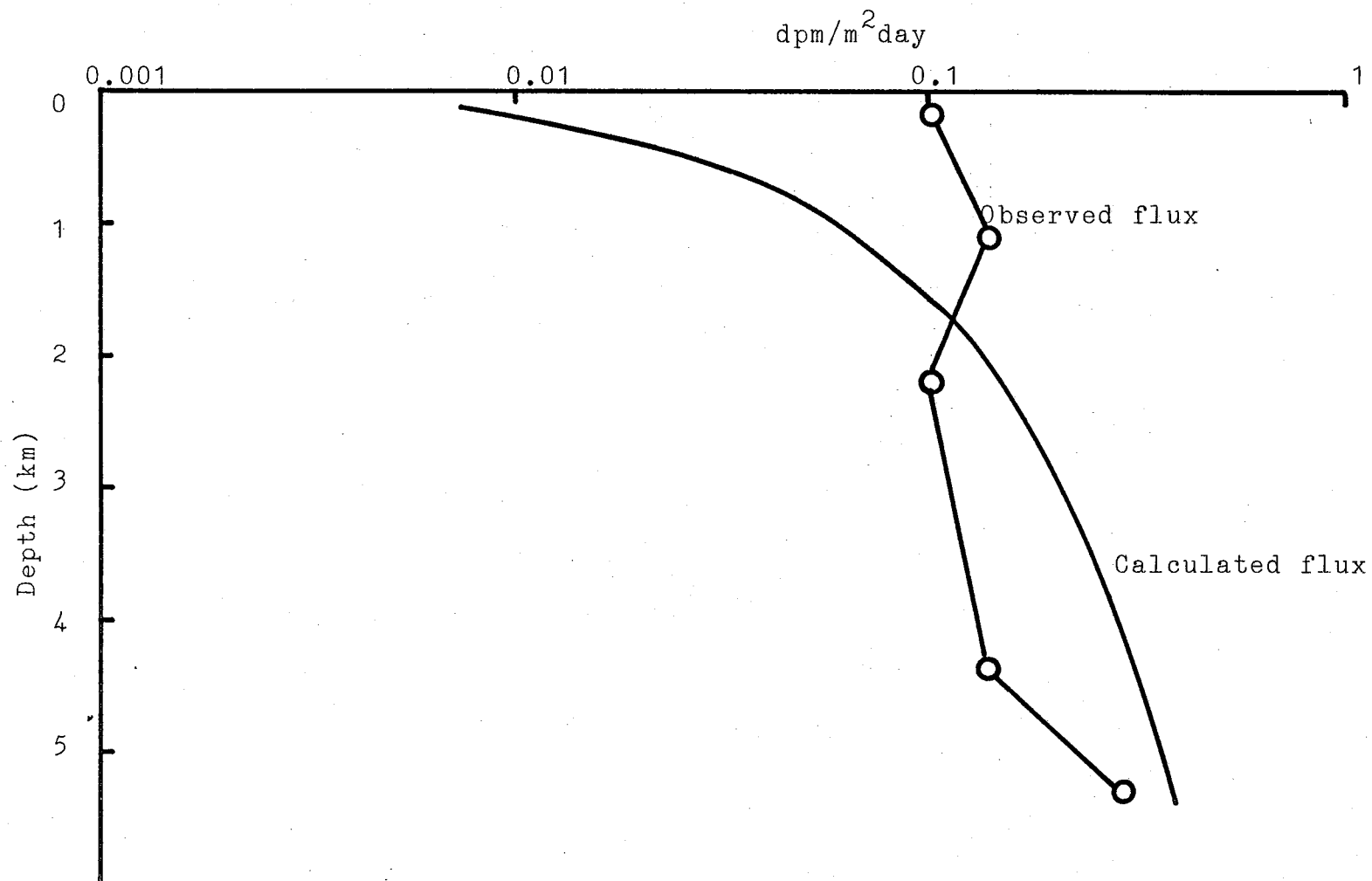


Figure III - 12 Flux of Th-230 observed with sediment traps in North Pacific and its net removal flux calculated.



CHAPTER IV CONCLUSIONS

1. The residence time of Be-7 in Funka Bay is found out to be 90-160 days, Which is nearly equal to that of Th-234.
2. The vertical distribution of Be-7 in the surface ocean is strongly effected by the particulate transport. The eddy diffusion coefficient of the surface water estimated from its vertical profile in the previous studies may not be real, because they have not taken its particulate transport into consideration.
3. The sediment trap experiments do not give net removal fluxes of chemical substances in sea water, but give downward fluxes of them.
4. The heavy metals in seawater are significantly transported downward by the particles which easily grow up to larger aggregates sinking with a significant speed and break down into smaller colloidal particles having negligibly small sinking velocity during their life in seawater.
5. A considerable amount of particulate matter moves up in the oceans together with water movement. The "Upward flux" of particulate matter seems to occur ordinarily everywhere in the oceans.

References:

- R. C. Aller and J. K. Cochran, $^{234}\text{Th}/^{238}\text{U}$ disequilibrium in near-shore sediment: particle reworking and diagenetic time scales, *Earth Planet. Sci. Lett.* 29 (1976) 37.
- K. Aoyagi, Geochemical study of ^{210}Pb and ^{210}Po in sea waters and sediments, B. Fish. Thesis, Hokkaido University (1975).
- T. M. Beasley, M. Heyrand, T. T. W. Higgo, R. D. Cherry and S. W. Fowler, ^{210}Po and ^{210}Pb in zooplankton fecal pellets, *Mar. Biol.* 44 (1978) 325.
- L. K. Benninger, The uranium-series radionuclides as tracers of geochemical processes in Long Island Sound, Ph. D. Thesis, Yale University, New Haven, (1976).
- S. G. Bhat, S. Krishnaswami, Rama and W. S. Moor, $^{234}\text{Th}/^{238}\text{U}$ ratios in the ocean, *Earth Planet. Sci. Lett.* 5 (1969) 483.
- J. K. B. Bishop, J. M. Edmond, D. R. Ketten, M. P. Bacon and W. B. Silker, The chemistry, biology, and vertical flux of particulate matter from the upper 400 m of the equatorial Atlantic Ocean, *Deep-Sea Res.* 24 (1977) 511.

P. G. Brewer, Y. Nozaki, W. D. Spencer and A. P. Fleer, A sediment trap experiment in the deep subtropical Atlantic; Isotopic and elemental fluxes, J. Mar. Res. 38 (1980) 703.

W. S. Broecker, A. Kaufman and R. M. Teier, The residence time of thorium in surface sea water and its implications, regarding the fate of reactive pollutants, Earth Planet. Sci. Lett. 20 (1973) 35.

G. Cozaz, E. Picciotto and W. De Breuck, Antarctic snow chronology with Pb-210, J. Geophys. Res. 69 (1964) 2597.

K. Fukuda and S. Tsunogai, A study of transport of continental aerosols by using ^{210}Pb in precipitation, Tellus 27 (1975) 514.

W. D. Gardner, Sediment trap dynamics and calibration: a laboratory evaluation, J. Mar. Res. 38 (1980) 17.

W. F. Graham, M. L. Bender and G. P. Klinkhammer, Manganese in Narragansett Bay, Limnol. Oceanogr. 21 (1976) 665.

K. Hirakawa and T. Kawamura, Seasonal changes of species of zooplankton in Funka Bay, in: Interim report of study on the oceanography of Funka Bay, T. Kawamura, Eds. Hokkaido University, Hakodate (1977).

S. Honjo, Material fluxes and modes of sedimentation in the mesopelagic and bathyplagic zones, Marine Res. 38 (1980) 53.

- C. E. Junge, Air chemistry and radioactivity (New York and London, Academic Press), (1963).
- D. P. Kharkar, J. Thomson, K. K. Turekian and W. O. Forster, Uranium and Thorium decay series nuclides in plankton from the Caribbean, *Limnol. Oceanogr.* 21 (1976) 269.
- M. Koide, A. Soutar and E. D. Goldberg, Marine geochronology with Pb-210, *Earth Planet. Sci. Lett.* 14 (1972) 442.
- M. Koide, K. W. Bruland and E. D. Goldberg, Th-228/Th-232 and Pb-210 geochronologies in marine and lake sediments, *Geoch. et Cosmoch. Acta* 37 (1973) 1171.
- D. Lal, On the investigations of geophysical processes using cosmic ray produced radioactivity, in: *Earth Science and Meteoritics* (North-Holland, Amsterdam), J. Geiss and E. D. Goldberg, Eds. (1963) 115.
- D. Lal and A. Lerman, Dissolution and behavior of particulate biogenic matter in the ocean: some theoretical considerations, 78 (1973) 7100.
- D. Lal and B. Peters, Cosmic ray produced radioactivity on the earth (SPRINGER-VerLag), S. Flugge Freiburg, Eds. Volume XLV1/2 (1967) 551.
- E. Matsumoto, ^{210}Pb geochronology of sediments from Lake Shinji, *Geochem. J.* 9 (1975) 167.

E. Matsumoto, ^{234}Th - ^{238}U radioactive disequilibrium in the surface layer of the ocean, *Geoch. et Cosmoch. Acta* 39 (1975) 205.

J. M. Merrill, E. F. X. Lyden, M. Honda and J. R. Arnold, The sedimentary geochemistry of the beryllium isotopes, *Geoch. et Cosmoch. Acta* 18 (1960) 108.

M. Minagawa and S. Tsunogai, Removal of ^{234}Th from a coastal sea: Funka Bay, Japan, *Earth Planet. Sci. Lett.* 47 (1980) 51.

Y. Nozaki and S. Tsunogai, ^{226}Ra , ^{210}Pb and ^{210}Po disequilibria in the Western North Pacific, *Earth Planet. Sci. Lett.* 32 (1976) 313.

K. Ohtani and K. Kido, Oceanographic structure in Funka Bay, *Bull. Fac. Fish. Hokkaido Univ.* 31 (1980) 84.

B. Peters, Cosmic rays, in: *Handbook of Physics* (McGraw-Hill Book Co.), Condon Odishaw, Eds. (1958) 201.

J. A. Robbins and D. N. Eginton, Determination of recent sedimentation rates in Lake Michigan using Pb-210 and Cs-137, *Geoch. et Cosmoch. Acta* 39 (1975) 285.

P. H. Santchi, Y. H. Li and J. Bell, Natural radionuclides in the water of Narragansett Bay, *Earth Planet. Sci. Lett.* 45 (1979) 201.

L. V. Shannon, R. D. Cherry and M. J. Orren, Polonium-210 and lead-210 in the marine environment, *Geoch. et Cosmoch. Acta* 34 (1970) 701.

T. Shinagawa, Atmospheric transport of chemical substances of land origin to the ocean with special reference to their impacts on the marine environment, Ph.D Thesis, Hokkaido University (1980) 193.

W. B. Silker, J. A. Young and M. R. Petersen, Oceanic distributions and relationship of ^7Be and fission products, *Radioactive Contamination of the Marine Environment*, IAEA-SM-158/46 (1973) 687.

D. W. Spencer, P. G. Brewer, A. P. Fleer, S. Honjo, S. Krishnaswami and Y. Nozaki, Chemical fluxes from a sediment trap experiment in the deep Sargasso Sea, *J. Mar. Res.* 36 (1978) 493.

S. Tsunogai, N. Tanaka and M. Uematsu, The relationship between particulate flux and sedimentation rate by the Pb-210 method in coastal region, paper presented at the annual meeting of Geochem. Soc. Japan, Tokyo, Oct. (1977)

S. Tsunogai, S. Noriki, N. Tanaka and M. Uematsu, New sediment trap and total particulate flux observed it, paper presented at the meeting of Oceanogr. Soc. Japan, Nagoya, Oct. (1980)

- S. Tsunogai and M. Minagawa, Settling model for the removal of insoluble chemical elements in sea water, *Geochem. J.* 12 (1978) 47.
- S. Tsunogai and N. Tanaka, Flux of oxygen across the sea-air interface as determined by the analysis of dissolved components in sea water, *Geochem. J.* 14 (1980) 227.
- S. Tsunogai and M. Uematsu, Particulate manganese, iron and aluminum in coastal water, Funka Bay, Japan, *Geochem. J.* 12 (1978) 39.
- S. Tsunogai, M. Uematsu, N. Tanaka, K. Harada, E. Tanoue and N. Handa, A sediment trap experiment in Funka Bay, Japan: "Upward flux" of particulate matter in sea water, *Mar. Chem.* 9 (1980) 321.
- S. Tsunogai and M. Yamada, A Radiochemically studied sediment core from the Philippine Sea Basin indicating no accumulation during the past few hundred thousand years, *Geochem. J.* 14 (1980) 19.
- K. K. Turekian, The fate of metals in the ocean, *Geoch. et Cosmoch. Acta* 41 (1977) 1139.
- L. V. Shannon, R. D. Cherry and M. J. Orren, Polonium-210 and lead-210 in the marine environment, *Geoch. et Cosmoch. Acta* 34 (1970) 701
- J. A. Young and W. B. Silker, Project BOMEX studies of atmospheric and oceanic mixing and air-sea exchange using radioactive tracer, *J. Geophys. Res.* 79 (1974) 4481.

APPENDIX I

Sedimentation rate of Pb-210 in Funka Bay.

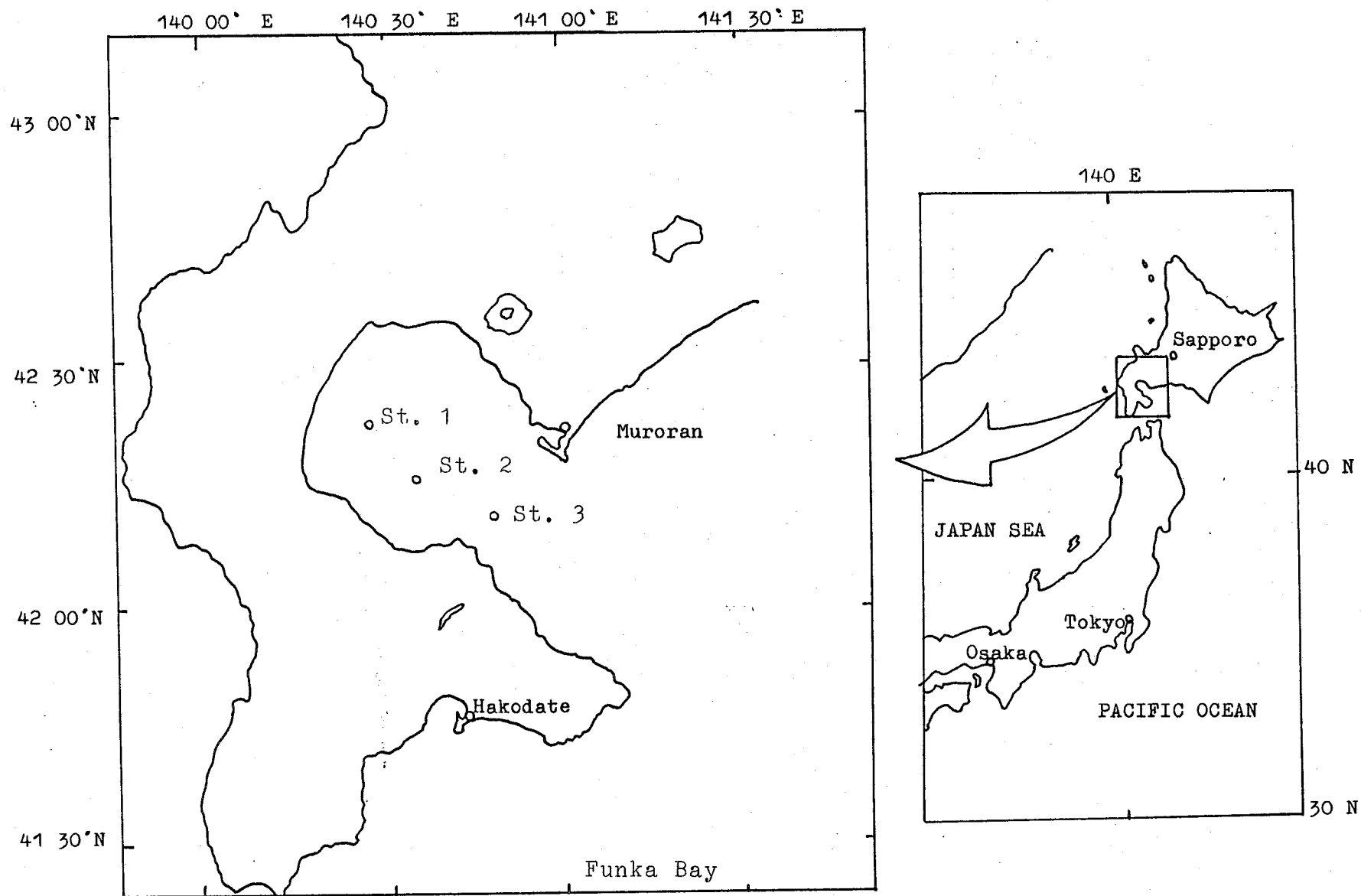


Figure 1. Sampling sites

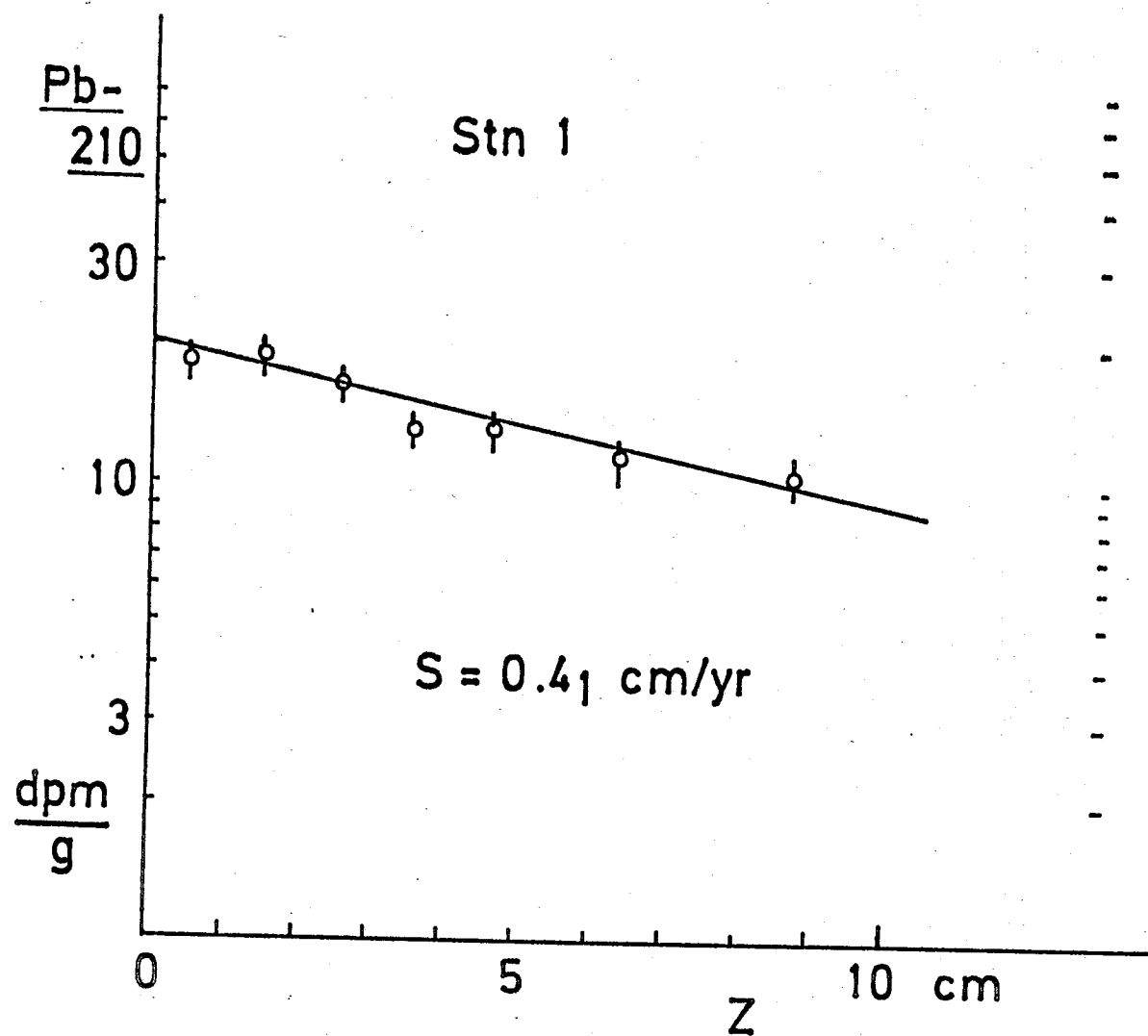


Figure 2a Vertical profile of Pb-210 in sediment at St. 1 in Funka Bay.

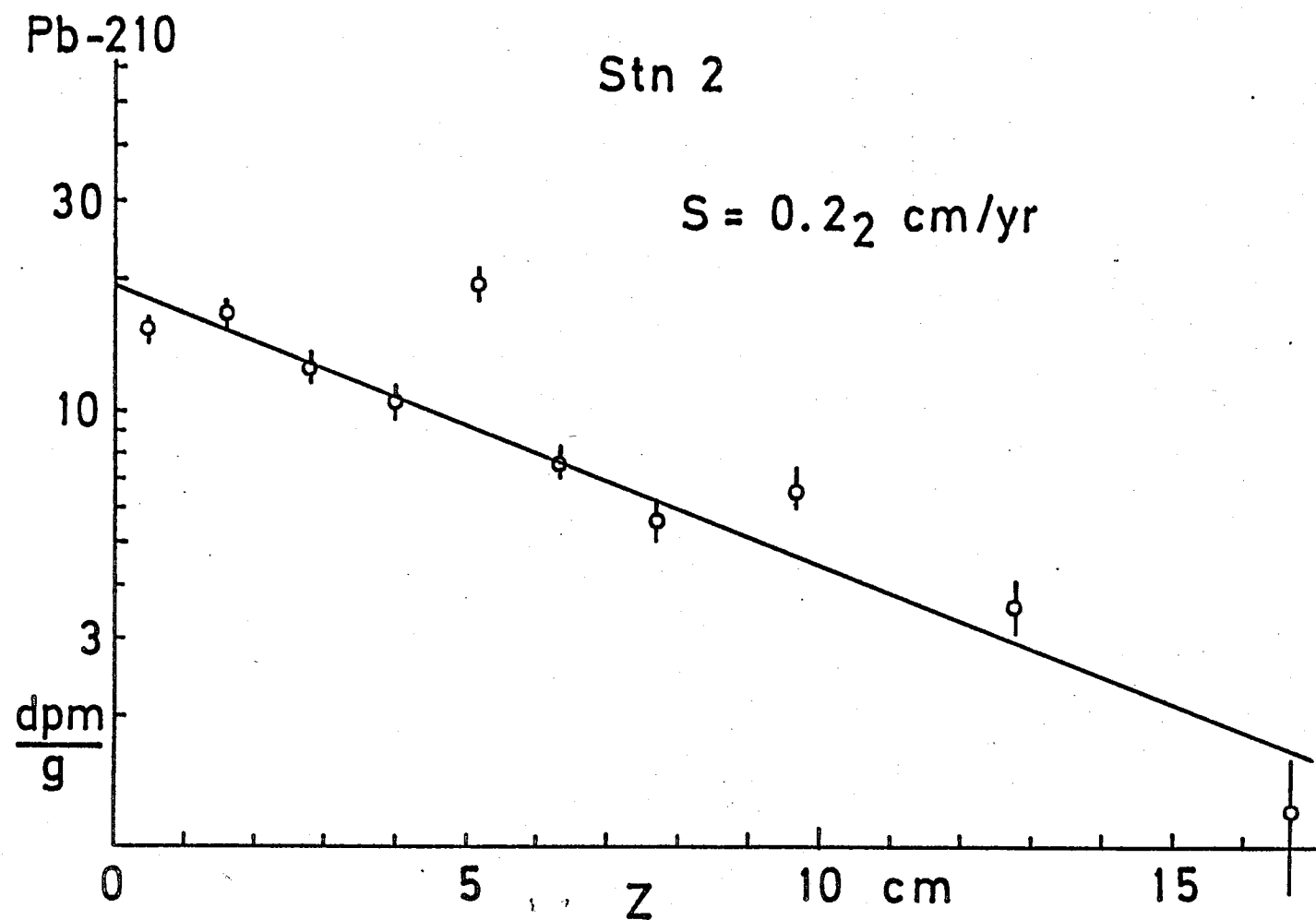


Figure 2b. Vertical distribution of Pb-210 in sediment at St. 2 in Funka Bay.

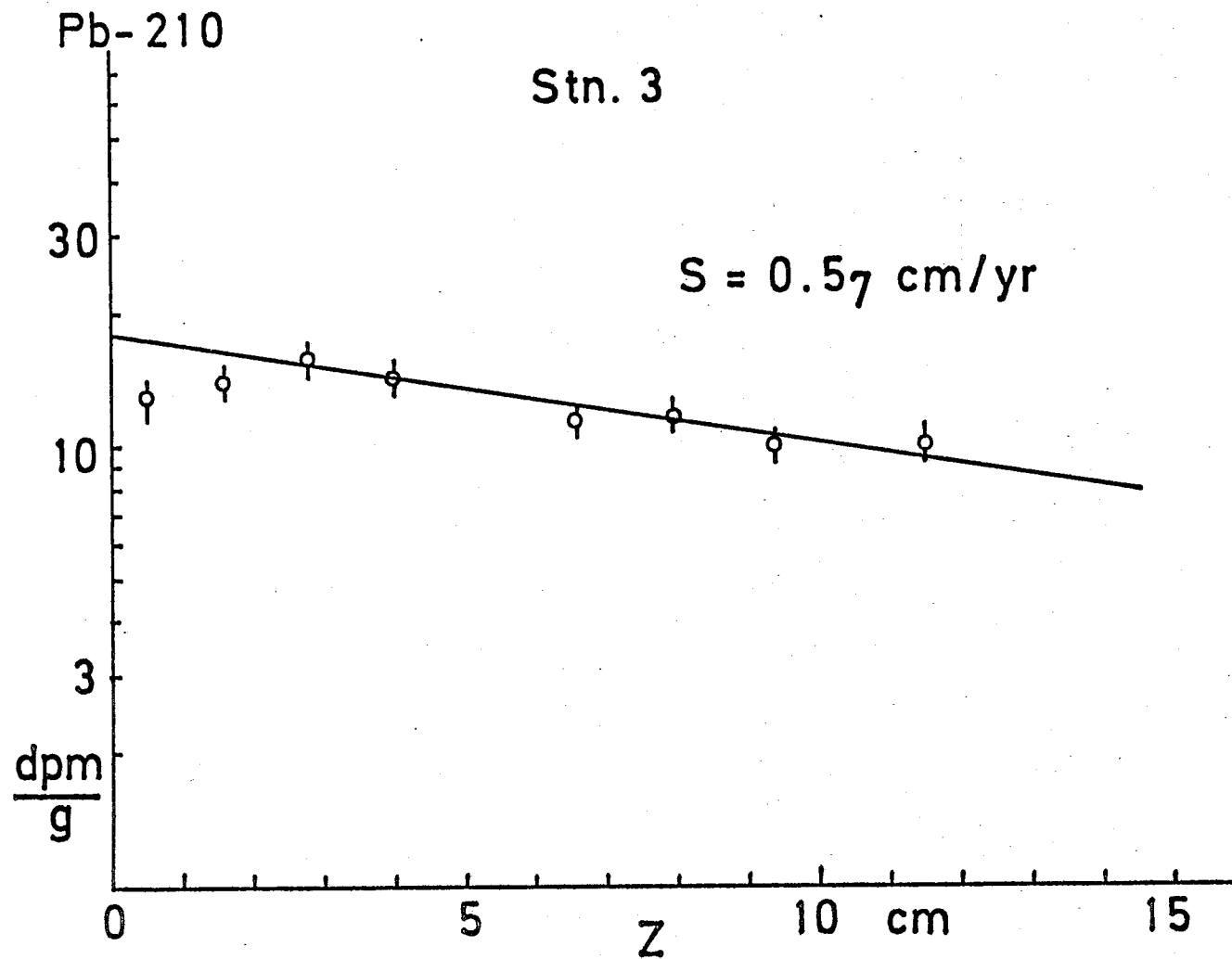


Figure 2c. Vertical distribution of Pb-210 in sediment at St. 3 in Funka Bay.

Table 1.

Relevant details on sampling sites and sediments collected.

Station	Location	Depth of water (m)	Description of samples studied
Stn.1	42 21.4'N 140 35.6'E	97	0-9.5cm dark green
Stn.2	42 17.8'N 140 35.3'E	92	0-14.0cm dark green
Stn.3	42 11.9'N 140 40.5'E	93	0-13.0cm dark green

Table 2.

 ^{210}Pb , porosity, pumice in Funka Bay

Depth interval (cm)	pumice (wt.%)	porosity	^{210}Pb (dpm/g)
Stn.1			
0-1	3.8	0.848	18.4 ± 1.4
1-2	3.3	0.843	18.9 ± 1.4
2-3	3.4	0.838	16.4 ± 1.2
3-4	4.3	0.835	13.3 ± 1.0
4-5	1.1	0.831	13.3 ± 1.0
5-7	0.0	0.833	11.4 ± 0.9
7-9.5	5.7	0.841	10.4 ± 0.8
Stn.2			
0-1	9.3	0.830	15.3 ± 1.1
1-2	5.9	0.804	16.5 ± 1.2
2-3	13.0	0.798	12.5 ± 1.0
3-4	14.0	0.794	10.3 ± 0.8
4-5	16.3	0.791	-
5-6	24.7	0.790	7.6 ± 0.6
6-7	39.8	0.779	5.6 ± 0.6
7-9	35.9	0.764	6.7 ± 0.7
9-11	50.3	0.726	3.6 ± 0.5
11-14	59.0	0.713	1.2 ± 0.4
85-90	-	-	0.6 ± 0.3
Stn.3			
0-1	3.8	0.829	13.1 ± 0.9
1-2	4.1	0.799	14.1 ± 1.0
2-3	6.7	0.793	15.8 ± 1.1
3-4	3.2	0.783	14.3 ± 1.0
4-5	5.8	0.775	-
5-6	3.8	0.770	11.4 ± 0.9
6-7	3.4	0.765	11.8 ± 0.9
7-8	1.3	0.767	10.0 ± 0.9
8-10	1.9	0.755	10.2 ± 0.9

Table 3 Sedimentation rate , porosity and input rate of Pb-210 to sediment.

Stn	Porosity (v/v)	Accumulation rate		
		S_o (cm/yr)	W_o (g/cm ² yr)*1	P (dpm/cm ² yr)
1	0.85	0.4 ₁	0.16	3.2
2	0.83	0.2 ₂	0.10	1.9
3	0.83	0.5 ₇	0.25	4.5

*1 calculated from the equation as follows :

$$W_o = \rho_s S_o (1 - \phi_o)$$

The meanings of variables are shown in text.

Table 4. The input rates to the sediments obtained from the $^{210}\text{Pb}_{\text{exc}}$ profiles measured in Funka Bay.

Sampling station	Input rate (dpm/cm ² yr)	References
1	(3.2) 0.43	this work Matsumoto (1980)
2	1.17 0.86	this work Nozaki (1977)
3	(4.5)	this work
45p	1.27	Matsumoto (1980)
82p	1.15	Matsumoto (1980)
85p	1.59	Matsumoto (1980)
19	2.02	Matsumoto (1980)

(): These values might include large errors, because $^{210}\text{Pb}_{\text{exc}}$ at the layer where it enough decays out was not available.

APPENDIX II

Hydrographic data in Funka Bay

Particulate flux* in Funka Bay observed with U type sediment traps. (St. 30)

Date	Depth (m)	Flux ($\mu\text{g}/\text{cm}^2\text{day}$)
1/13-1/18 1978	1	767
	40	1634
	60	-
	80	1637
	85	-
5/16-5/19 1978	1	53.5
	40	62.1
	60	77.5
	80	113.6
	85	179.2
6/17-6/22 1978	1	-
	40	79.6
	60	105.3
	80	131.4
	85	106.9
8/23-8/26 1978	1	45.2
	40	126.5
	60	64.8
	80	155.0
	85	297.0
11/25-11/30 1978	1	-
	40	-
	60	944
	80	925
	85	1346

(continued)

Date	Depth (m)	Flux ($\mu\text{g}/\text{cm}^2\text{day}$)
12/14-12/16 1978	1	209
	40	227
	60	308
	80	364
	85	-

Date	Depth Flux		Depth Flux		
	(m)	($\mu\text{g}/\text{cm}^2 \cdot \text{day}$)	(m)	($\mu\text{g}/\text{cm}^2 \cdot \text{day}$)	
	[Station 30]		[Station 2]		
4/23- 4/25 1979	1	365 $\pm$ 163 [†]	1	129 $\pm$ 48	
	40	179 $\pm$ 26	30	138 $\pm$ 16	
	60	208 $\pm$ 11	50	208 $\pm$ 10	
	80	251 $\pm$ 18	80	346 $\pm$ 21	
5/31- 6/ 1 1979	1	119 $\pm$ 17	1	74 $\pm$ 49	
	40	139 $\pm$ 27	30	125 $\pm$ 14	
	60	111 $\pm$ 5	50	139 $\pm$ 29	
	80	275 $\pm$ 82	80	189 $\pm$ 39	
6/25- 6/27 1979	1	102 $\pm$ 17	1	98 $\pm$ 8	
	40	239 $\pm$ 31	30	346 $\pm$ 24	* Mean of values obtained from 6 cylinders standard deviations. (one sigma)
	60	59 $\pm$ 15	50	67 $\pm$ 32	
	80	179 $\pm$ 45	80	87 $\pm$ 16	
7/16- 7/18 1979	1	111 $\pm$ 40 [†]	1	146 $\pm$ 29	
	40	56 $\pm$ 21 [†]	30	69 $\pm$ 13	† Contained some living zooplankton.
	60	78 $\pm$ 20 [†]	50	120 $\pm$ 17	
	80	76 $\pm$ 1 [†]	80	-	
8/20- 8/22 1979	1	53 $\pm$ 8	1	67 $\pm$ 25	‡ Rough weather at the retrieval.
	40	231 $\pm$ 71	30	158 $\pm$ 8	
	60	88 $\pm$ 6	50	135 $\pm$ 24	
	80	230 $\pm$ 27	80	250 $\pm$ 23	
9/17- 9/19 1979	1	29 $\pm$ 18	1	46 $\pm$ 7	§ Dec 10-13, 1979
	40	103 $\pm$ 16	30	115 $\pm$ 10	
	60	169 $\pm$ 21	50	72 $\pm$ 4	
	80	156 $\pm$ 36	80	178 $\pm$ 15	
11/20-11/22 1979	1	263 $\pm$ 24	1	143 $\pm$ 13	¶ Feb 25-28, 1980
	40	268 $\pm$ 21	30	146 $\pm$ 6	
	60	305 $\pm$ 26	50	160 $\pm$ 7	
	80	468 $\pm$ 21	80	447 $\pm$ 37	
12/12-12/13 1979	1	373 $\pm$ 40	1	137 $\pm$ 13§	
	40	165 $\pm$ 17	30	169 $\pm$ 10§	
	60	156 $\pm$ 19	50	156 $\pm$ 13§	
	80	402 $\pm$ 25	80	350 $\pm$ 28§	
1/23- 1/28 1980	1	451 $\pm$ 138	1	173 $\pm$ 11	
	40	956 $\pm$ 7	30	228 $\pm$ 14	
	60	1049 $\pm$ 16	50	259 $\pm$ 8	
	80	1518 $\pm$ 61	80	418 $\pm$ 40	
2/26- 2/28 1980	1	235 $\pm$ 31	1	250 $\pm$ 38¶	
	40	252 $\pm$ 17	30	618 $\pm$ 50¶	
	60	380 $\pm$ 29	50	860 $\pm$ 68¶	
	80	781 $\pm$ 28	80	1716 $\pm$ 137¶	
3/14- 3/15 1980	1	347 $\pm$ 151	1	59 $\pm$ 34 †	
	40	199 $\pm$ 42	30	165 $\pm$ 20	
	60	305 $\pm$ 23	50	508 $\pm$ 90	
	80	1181 $\pm$ 90	80	688 $\pm$ 65	

[UP-79-4]

Sta. 2. Apr. 25, 1979. 17:52

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄
0	3.3	32.507	8.97	0.43	5.0	0.30
18.2	1.57	32.565	8.83	-	3.5	0.49
25.6	-	-	-	1.68	-	-
32.5	1.43	32.725	8.29	-	11.2	0.99
43.9	-	-	-	1.07	-	-
51.6	1.31	32.871	7.64	-	18.6	1.40
61.2	1.49	32.902	7.28	-	27.4	1.58
62.1	-	-	-	2.00	-	-
70.8	1.50	32.904	7.13	-	27.8	1.67
80.3	1.61	32.928	7.17	0.47	22.8	1.55
89.9	2.84	33.408	6.13	-	35.0	1.63

Sta. 30. Apr. 25, 1979. 09:43

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄
0	4.2	32.374	7.91	0.40	7.5	0.42
15.0	2.61	32.562	9.30	-	4.6	0.36
20.0	-	-	-	0.71	-	-
30.0	2.17	32.611	8.75	-	3.7	0.41
40.0	-	-	-	1.32	-	-
50.0	2.09	32.763	7.96	-	5.0	0.85
60.0	1.99	32.826	7.73	0.69	10.4	1.09
70.0	2.90	33.338	6.34	-	23.6	1.55
80.0	-	-	-	1.08	-	-
90.0	3.35	33.599	5.53	-	40.6	1.52

[UP-79-5]

Sta. 30, June 1. 1979.

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄
0	9.2	32.027	6.96	1.36	8.0	0.18
15.0	8.84	32.041	6.99	-	5.8	0.19
19.9	8.56	-	-	1.15	-	-
29.9	4.14	32.410	7.68	-	3.4	0.37
39.8	3.81	-	-	0.80	-	-
49.8	2.77	32.632	7.65	-	8.1	0.59
57.8	2.30	-	-	0.56	-	-
61.8	2.27	32.746	7.44	-	7.8	0.94
69.7	2.16	32.819	7.01	-	14.7	1.22
77.7	2.60	-	-	1.51	-	-
81.7	2.63	33.144	6.34	-	35.3	1.59
89.7	2.64	33.150	6.31	-	37.6	1.53

Sta. 30, June 1. 1979.

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄
20.0	8.84	-	-	-	-	-
40.0	4.24	-	-	-	-	-
60.0	2.40	-	-	-	-	-
78.0	2.53	-	-	-	-	-
80.0	2.76	33.186	6.87	-	36.9	1.54
82.0	2.76	33.191	6.38	-	37.9	1.55
84.0	2.78	33.191	6.12	-	35.3	1.51
86.3	2.76	33.188	6.12	-	35.5	1.56
88.0	2.73	33.191	6.10	-	35.1	1.59
90.0	2.75	33.192	6.08	-	35.5	1.73
92.0	2.73	33.175	5.82	-	-	-

[UP-79-6]

Sta. 2, June 27. 1979. 14:25

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (µg at./l)	PO ₄
0	14.7	31.756	6.39	-	2.9	0.12
20.5	6.39	32.989	8.23	-	7.4	0.45
40.9	2.70	32.913	7.57	-	25.0	1.35
57.3	4.46	33.282	7.05	-	18.7	1.09
65.5	4.03	33.323	6.69	-	23.7	1.36
77.8	4.27	33.453	6.28	-	29.1	1.31

Sta. 2, June 27. 1979. 15:00

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (µg at./l)	PO ₄
32.7	3.07	32.802	7.92	3.54	13.1	0.98
49.1	3.63	33.075	7.38	1.39	22.6	1.14
73.7	4.09	33.368	6.40	0.79	27.5	1.29
0	-	-	-	0.78	-	-

Sta. 30, June 27. 1979. 10:51

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (µg at./l)	PO ₄ (µg at./l)	NO ₂	NO ₃
0	14.6	31.829	6.33	0.77	10.5	0.06	0	0.32
17.4	14.10	31.869	6.40	-	2.8	0.03	0	0
31.9	5.43	32.381	7.63	-	12.0	0.03	0	0
51.2	2.39	32.768	6.83	-	14.1	1.09	0.13	4.01
62.8	2.43	32.941	6.60	-	24.0	1.37	0.31	7.68
70.5	2.76	33.042	6.60	-	27.1	1.35	0.36	11.45
82.1	3.03	33.153	6.21	-	29.1	1.49	0.69	8.98
89.8	3.61	33.294	5.96	-	40.1	1.50	0.90	11.97
19.3	-	-	-	1.03	-	-	-	-
38.6	-	-	-	0.75	-	-	-	-
56.0	-	-	-	1.11	-	-	-	-
75.3	-	-	-	1.22	-	-	-	-

[UP-79-7]

Sta. 2, July 16. 1979. 09:08

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (µg at./l)	PO ₄
0	15.6	32.198	6.27	-	-	0.01
14.9	11.39	32.757	7.33	-	5.1	0.21
29.8	-	33.433	7.76	-	4.0	0.44
39.8	7.16	33.649	6.70	-	13.0	0.76
49.7	-	33.762	6.42	1.38	9.9	0.85
59.7	6.80	33.667	6.29	-	11.5	0.91
69.6	6.58	33.647	6.32	-	14.2	0.96
79.6	-	33.685	5.79	2.36	13.7	0.84
84.5	6.32	33.672	5.67	-	23.3	1.07
89.5	6.09	33.643	5.66	-	23.1	1.05
92.5	6.08	33.637	5.62	-	20.0	1.03

Sta. 30, July 16, 1979. 16:14

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄
0	(16.4)	31.591	6.57	0.94	0.7	0.10
20.0	11.92	32.412	6.92	-	1.7	0.20
24.7	10.40	-	-	1.61	-	-
34.1	4.34	32.482	7.66	-	3.0	0.47
43.4	3.20	-	-	1.61	-	-
52.7	3.74	32.939	6.89	-	11.9	1.06
60.2	4.12	-	-	1.03	-	-
64.0	4.45	33.170	6.64	-	17.4	1.16
71.4	2.79	33.216	5.50	-	24.4	1.56
78.9	3.22	-	-	1.49	-	-
82.6	3.25	33.275	4.70	-	52.6	1.60
90.1	3.00	33.297	2.55	-	64.7	2.07

Sta. 30, July 16, 1979. 15:45

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄
0	-	-	5.85	-	-	-
10.0	-	31.991	6.01	-	-	-
30.0	-	32.427	7.62	-	-	-
50.0	-	32.899	7.08	-	-	-
80.0	-	32.933	6.64	-	-	-
82.0	-	33.066	6.13	-	20.2	1.21
84.0	-	33.031	6.18	-	20.7	1.30
86.0	-	33.056	5.93	-	20.0	1.42
88.0	-	33.091	5.84	-	23.4	1.35
90.0	-	33.085	5.67	-	28.8	1.47

Sta. 30, July 17, 1979. 09:00~10:00

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄ $\mu\text{g at.}/\text{l}$	NO ₂	NO ₃
0	16.4	-	-	-	-	-	0	0
20.0	-	-	-	-	-	-	0	0
40.0	-	-	-	-	-	-	0.02	1.75
60.0	-	-	-	-	-	-	0.56	6.82
80.0	-	-	-	-	-	-	0.26	13.79
82.0	3.31	-	-	-	-	-	0	18.42
84.0	3.66	-	-	-	-	-	0	18.86
86.0	3.66	-	-	-	-	-	0.01	13.06
88.0	3.61	-	-	-	-	-	0.03	15.87
90.0	3.46	-	-	-	-	-	0	15.83
92.0	3.23	-	-	-	-	-	0.25	15.48

[UP-79-8]

Sta. 2, Aug. 20, 1979. 09:55

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at./l}$)	PO ₄
0	21.4	33.173	5.14	0.618	1.5	0.05
22.8	16.67	33.828	6.04	-	5.5	0.11
36.5	12.37	33.888	6.57	1.053	5.4	0.16
45.7	11.16	33.976	6.30	-	5.4	0.33
54.8	10.44	34.078	5.98	0.844	6.2	0.46
63.9	9.60	34.007	5.51	-	5.9	0.56
73.1	9.25	34.004	5.73	-	4.2	0.61
82.2	8.78	33.960	5.35	0.988	14.5	0.88
86.8	8.76	33.964	5.32	-	19.7	0.92
91.4	8.71	33.957	5.23	-	21.2	0.92
94.1	8.64	33.945	5.27	-	36.6	0.98

Sta. 30, Aug. 20, 1979. 16:40

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at./l}$)	PO ₄
0	21.5	31.926	5.08	0.821	4.0	0.09
15.0	20.94	32.417	5.14	-	4.4	0.08
20.0	-	-	-	-	-	-
30.0	12.67	32.710	6.68	-	9.4	0.19
40.0	-	-	-	0.706	-	-
50.0	3.39	32.668	7.16	-	4.7	0.76
58.0	-	-	-	1.013	-	-
62.0	2.96	32.863	6.24	-	15.2	1.25
70.0	-	33.025	5.60	-	22.0	1.34
78.0	-	-	-	1.145	-	-
82.0	-	33.261	2.72	-	65.4	1.88
90.0	-	33.280	2.16	-	73.4	2.20

Sta. 30, Aug. 21, 1979. 12:25

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at./l}$)	PO ₄ $\mu\text{g at./l}$	NO ₂	NO ₃
77.0	2.99	33.168	5.04	-	-	-	-	-
78.8	3.38	33.234	3.87	-	-	-	0	17.33
80.7	3.24	33.265	2.93	-	-	-	0.02	20.11
82.5	3.26	33.271	2.53	-	-	-	0	19.28
84.3	3.21	33.280	-	-	-	-	0	20.08
86.2	3.25	33.279	2.33	-	-	-	0	19.28
88.1	3.25	33.280	2.39	-	-	-	0	20.16

Sta. 30, Aug. 21, 1979. 15:55

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄ $\mu\text{g at.}/\text{l}$	NO ₂	NO ₃
0	-	-	-	-	-	-	0	0
24.9	-	-	-	-	-	-	0	0
43.3	-	-	-	-	-	-	0	0
61.7	-	-	-	-	-	-	0.11	1.05
80.1	-	-	-	-	-	-	0.25	7.45
81.9	-	-	-	-	-	-	0	14.76
83.8	-	-	-	-	-	-	0	16.69
85.6	-	-	-	-	-	-	0	16.58
87.4	-	-	-	-	-	-	0	18.49
89.3	-	-	-	-	-	-	0	11.49
91.1	-	-	-	-	-	-	0	19.71

[UP-79-9]

Sta. 2, Sept. 17, 1979. 09:00

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄ $\mu\text{g at.}/\text{l}$
0	19.1	33.173	5.47	0.464	8.2	0.05
14.5	16.54	33.828	5.93	-	9.2	0.15
38.6	13.21	33.888	5.52	0.493	13.7	0.50
58.0	12.42	33.996	5.43	0.481	19.5	0.61
67.6	6.79	34.078	5.16	-	30.7	1.17
82.1	6.26	34.009	5.17	0.619	34.9	1.23
86.9	6.08	34.004	5.17	-	36.9	1.32
89.8	6.20	33.960	5.18	-	36.4	1.30

Sta. 30, Sept. 17, 1979. 14:40

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at.}/\text{l}$)	PO ₄ $\mu\text{g at.}/\text{l}$
0	19.8	32.651	6.35	0.634	7.2	0.04
15.0	18.97	33.201	5.73	-	5.1	0.04
30.0	10.09	32.806	7.22	-	5.5	0.29
50.0	10.00	32.775	5.67	-	21.6	0.80
60.0	9.49	32.925	5.65	0.630	28.0	0.94
70.0	6.82	33.119	5.75	-	23.6	1.01
80.0	4.55	33.278	4.542	0.885	49.5	1.35
90.0	3.35	33.281	2.26	-	76.2	1.88
20.0	-	-	-	0.627	-	-
40.0	-	-	-	0.543	-	-

Sta. 30, Sept. 18, 1979. 12:04

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (	PO ₄ μg at./l	NO ₂	NO ₃)
0	19.6	32.859	5.73	-	2.7	0.12	0	0
19.8	17.13	32.632	6.02	-	5.5	0.12	0	0
39.6	12.17	32.672	5.51	-	17.2	0.65	0.21	3.37
59.4	9.84	32.894	(5.66)	-	19.9	0.94	0.09	5.33
79.2	5.51	33.277	5.61	-	24.7	1.11	0	8.04
81.2	5.29	33.612	5.62	-	26.6	1.16	0.01	8.95
83.2	4.97	33.010	5.51	-	26.4	1.28	0	8.27
85.2	4.83	32.909	5.51	-	30.0	1.32	0.02	10.89
87.1	4.56	32.737	5.33	-	34.0	1.37	0	10.96
89.1	4.17	32.677	4.87	-	43.8	1.57	0.01	12.99
91.9	3.82	32.832	3.93	-	65.3	1.82	0.02	15.65

[UP-79-11]

Sta. 2, Nov. 20, 1979. 10:05

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (μg at./l)	PO ₄
0	13.0	-	5.58	-	9.5	0.56
10.0	12.90	-	5.40	-	10.0	0.74
20.0	12.85	-	5.68	-	10.4	0.43
30.0	12.82	-	5.58	-	9.7	0.37
40.0	12.81	-	5.49	-	8.6	0.29
50.0	12.81	-	5.49	-	9.7	0.35
60.0	12.79	-	5.55	-	9.7	0.31
70.0	12.05	-	5.46	-	13.1	0.47
80.0	11.03	-	5.09	-	21.5	0.94
90.0	10.98	-	5.10	-	22.7	0.95

Sta. 30, Nov. 21, 1979. 10:17

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (	PO ₄ μg at./l	NO ₂	NO ₃)
0	11.5	33.923	5.84	0.179	13.5	0.54	0.34	4.12
20.0	11.70	33.922	5.83	0.136	12.8	0.55	0.33	3.91
40.0	11.68	33.913	5.79	0.213	13.2	0.41	0.35	4.42
59.0	11.43	33.924	5.61	0.215	16.9	0.63	0.15	11.02
69.0	11.40	-	5.28	-	24.0	0.93	0.08	12.62
79.0	7.87	33.791	4.58	0.794	44.9	1.52	-	-
89.0	7.66	33.810	4.34	-	51.4	1.69	-	-

Sta. 30, Nov. 21, 1979.

Depth- (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (	PO ₄ μg at./l	NO ₂	NO ₃)
78.0	8.76	33.842	4.81	-	5.2	1.22	0	14.83
80.0	8.15	33.797	4.62	-	44.9	1.42	0	14.39
82.0	7.81	33.752	4.39	-	52.1	1.61	0.01	17.51
84.0	7.62	33.745	4.25	-	54.1	1.71	0.02	15.18
86.0	7.83	33.783	4.35	-	54.7	1.66	0.16	17.72
88.0	7.70	33.772	4.16	-	57.8	1.71	0.21	16.74
90.0	7.68	33.778	4.27	-	54.1	1.82	0.28	17.48

[UP-79-12]

Sta. 30, Dec. 12, 1979. 09:44

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (	PO ₄ μg at./l	NO ₂	NO ₃)
0	9.9	33.943	5.95	0.190	20.8	0.69	0.47	6.46
19.0	10.04	33.929	5.90	0.712	22.3	0.70	0.46	6.14
38.0	9.95	33.924	5.87	0.325	20.3	0.68	0.31	6.27
57.0	9.75	33.917	5.48	0.470	25.4	0.82	0.30	8.46
76.0	8.43	33.820	4.01	0.983	50.4	1.57	0.04	15.05
78.0	8.24	33.824	3.85	-	54.8	1.66	0	13.42
79.0	8.08	33.791	3.85	-	58.3	1.91	0.01	15.52
81.0	8.08	33.807	3.69	-	62.0	2.14	0.09	15.58
83.0	8.08	33.816	3.74	-	62.2	2.09	0.27	14.49
85.0	8.07	33.810	3.66	-	65.3	1.94	0.20	15.89
87.0	8.03	33.804	3.78	-	67.9	1.91	0.11	16.01

[UP-80-1]

Sta. 2, Jan. 23, 1980.

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (	PO ₄ μg at./l
0	7.4	33.777	6.37	-	28.1	0.96
14.9	7.67	33.770	6.32	-	25.3	0.90
29.7	7.65	33.769	6.30	-	26.6	0.95
49.5	7.72	33.767	6.34	-	25.6	0.89
59.4	7.72	33.795	6.24	-	23.9	0.95
69.3	8.12	33.907	-	-	24.8	0.99
79.2	8.11	33.943	6.24	-	23.4	0.87
89.1	7.75	33.959	6.26	-	19.8	1.03

Sta. 30, Jan. 28, 1980.

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (μ g at./l)	PO ₄ (μ g at./l)	NO ₂	NO ₃ (μ g at./l)
0	6.9	33.916	6.60	0.555	21.6	0.90	0.15	9.08
19.1	6.87	33.917	6.55	-	25.6	0.82	0.17	9.24
38.2	6.74	33.954	6.53	(1.769)	21.1	0.92	0.22	8.73
57.4	6.57	34.004	6.42	-	22.7	1.22	0.47	7.90
76.5	6.30	34.007	6.47	0.913	29.5	0.94	0.48	7.68
78.4	6.34	34.006	6.45	1.365	27.7	0.95	0.47	7.69
80.3	6.26	34.009	6.49	-	25.7	1.00	0.48	7.58
82.2	6.26	34.008	6.49	-	26.8	1.02	0.45	7.56
84.2	6.24	34.008	6.48	-	29.7	0.88	0.47	7.09
86.1	6.25	34.009	6.43	-	33.3	0.95	0.12	8.82
88.0	6.26	34.006	6.39	-	29.1	0.99	0.20	9.26

[UP-80-2]

Sta. 2, Feb. 25, 1980.

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (μ g at./l)	PO ₄ (μ g at./l)
0	4.9	33.660	6.75	0.437	31.1	1.01
15.0	5.20	33.624	6.73	-	31.9	0.99
29.9	5.24	33.629	6.76	0.870	27.7	0.99
49.8	5.08	33.673	6.90	0.789	24.3	0.99
59.9	4.92	33.703	6.99	-	29.1	0.93
69.8	4.69	33.846	6.90	-	41.1	0.85
79.8	4.59	33.916	6.93	0.915	26.5	0.87
89.8	4.54	33.918	6.90	-	27.9	0.87

Sta. 30, Feb. 26, 1980. 11:37

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si (μ g at./l)	PO ₄ (μ g at./l)	NO ₂	NO ₃ (μ g at./l)
0	4.2	33.663	7.42	0.445	25.4	0.81	0.24	7.15
19.2	-	33.736	7.23	0.970	38.3	0.80	0.23	8.20
38.5	4.57	33.899	6.99	0.671	29.1	0.79	0.30	7.68
57.7	4.56	33.932	6.96	0.898	22.7	0.79	0.33	7.42
76.9	4.59	33.930	6.94	1.365	31.0	0.83	0.36	7.08
78.8	4.53	33.931	6.90	-	28.9	0.83	0.33	7.31
80.7	4.52	33.934	6.91	-	27.7	0.82	0.37	6.57
82.7	4.51	33.936	6.91	-	23.5	0.81	0.37	6.76
84.6	4.50	33.934	6.92	-	30.7	0.87	0.34	6.16
86.5	4.47	33.934	6.93	-	29.0	0.83	0.32	6.52
88.4	4.54	33.947	6.79	-	31.3	0.81	0.31	6.21

[UP-80-3]

Sta. 2, Mar. 14, 1980. 10:57

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at./l}$)	PO ₄
0	5.8	33.680	6.79	0.316	22.8	0.86
20.0	5.73	33.681	6.72	-	-	0.85
40.0	5.75	33.685	6.72	-	22.5	0.86
60.0	6.07	33.784	6.39	-	18.9	0.87
70.0	6.13	33.800	6.36	-	21.6	0.87
80.0	6.15	33.800	6.35	-	21.5	0.87
84.0	6.16	33.806	6.37	-	22.3	0.86
32.2	-	-	-	0.348	-	-
50.6	-	-	-	0.386	-	-
78.2	-	-	-	0.441	-	-

Sta. 30, Mar. 15, 1980. 09:00

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at./l}$)	PO ₄ $\mu\text{g at./l}$	NO ₂	NO ₃
0	-	-	-	1.07	-	-	-	-
17.0	-	-	-	1.16	-	-	-	-
35.6	-	-	-	0.91	-	-	-	-
52.6	-	-	-	0.99	-	-	-	-
69.5	-	-	-	0.98	-	-	-	-
83.6	-	-	-	-	-	-	-	-
85.5	-	-	-	-	-	-	0.28	5.93
87.5	-	-	-	-	-	-	0.26	6.11
89.4	-	-	-	-	-	-	0.26	5.94
91.3	-	-	-	-	-	-	0.26	5.85
							0.27	5.75

Sta. 30, Mar. 15, 1980. 09:44

Depth (m)	Temp. (°C)	Sal. (‰)	D.O. (ml/l)	SS (mg/l)	Si ($\mu\text{g at./l}$)	PO ₄ $\mu\text{g at./l}$	NO ₂	NO ₃
0	(3.8)	33.771	7.53	-	16.6	0.50	0.17	2.56
23.1	3.94	33.765	7.58	-	16.6	0.56	0.16	2.57
42.3	4.00	33.775	7.40	-	19.0	0.57	0.19	3.73
61.5	4.02	33.778	7.39	-	18.5	0.66	0.18	4.41
80.9	4.10	33.838	7.18	-	20.2	0.65	0.21	4.19
82.7	4.23	33.837	7.12	-	19.2	0.68	-	-
84.5	4.23	33.846	7.13	-	19.1	0.64	-	-
86.5	4.14	33.865	7.07	-	23.1	0.73	-	-
88.4	4.15	33.881	6.98	-	22.8	0.83	-	-
90.3	4.18	33.885	6.96	-	28.0	0.85	-	-
92.0	4.19	33.878	6.71	-	-	-	-	-