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659. THORIUM AND PROTACTINIUM ISOTOPES IN SOME PRESENT-DAY HERMATYPIC CORALS AND THEIR IMPLICATIONS TO DATING*

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現生造礁性サンゴ中のトリウムおよびプロトアクチニウム同位体とそれらの年代測定への影響：四国室戸岬から New Britain 島にかけての太平洋西縁部に沿う9地域の現生造礁性サンゴを，U·Th および Pa 同位体組成と U の分布様式について検討した。その結果，ほとんどの試料で測定可能量の Th および Pa 同位体が検出された。また，南西諸島産化石試料中には，同地域の現生試料と同程度の ^{232}Th が含まれていることから，少なくとも南西諸島産化石サンゴから ^{230}Th および ^{231}Pa 放射年令を求める場合には，初生的な ^{230}Th と ^{231}Pa 量を見積り，補正年令を求める必要がある。さらに，フィッション・トラック法で観察された U の不均一分布は，^{238}U 量の部分的な差が最大 30% に達し，^{238}U 最多部と最少部間で見かけの ^{230}Th·^{231}Pa 年令ともに，計数誤差以上の差を生ずる原因になる可能性もある。見かけの ^{230}Th 年令値の補正は，現生種の ^{230}Th/^{232}Th 放射能比が 1.4～3.0 と限られた範囲に入ることから，化石試料中の ^{232}Th 量が求まれば，近似的には可能である。ところが，今回得られた現生種の ^{230}Th/^{232}Th 比が，生息域の海水の同比より，見かけ上いく分高い事に注目しなければならない。このことは，各試料の分析された部位が数年～数十年以前に形成された部分であるため，試料採集時までに直接の親核種である ^{234}U から成長した ^{230}Th が，骨格分泌当時の ^{230}Th/^{232}Th 比を，見かけ上増大させたためと説明される。結局，化石サンゴの見かけの ^{230}Th 年令の補正に用いる初期 ^{230}Th/^{232}Th 比として，試料産出地付近の海水の ^{230}Th/^{232}Th 比を用いる事が，もっとも適切な方法といえる。本小論では，南西諸島化石サンゴに，与論島および徳之島の沿岸水の ^{230}Th/^{232}Th 比の平均値 (1.4) を初期 ^{230}Th/^{232}Th 比として用い，補正 ^{230}Th 年令を求めたところ，矛盾のない値を得ることが出来た。

大 村 明 雄

Introduction

Both the deficient ^{230}Th and ^{231}Pa methods allow to extend the radiometric Pleistocene time scale back to about 250,000 years, and have greatly contributed to solve the Quaternary problems such as fluctuation of sea-level and tectonic movement (*e.g.* BROECKER *et al.*, 1968; MESOLELLA *et al.*, 1969; JAMES *et al.*, 1971; STEINEN *et al.*, 1973; VEEH and CHAPPELL, 1970; CHAPPELL, 1974a, 1974b; KONISHI *et al.*, 1970, 1974). Yet there is still a room to reexamine rigorously the reliability of ^{230}Th and ^{231}Pa dates of hermatypic corals, which are accepted as the most favorable sample lacking the postmortem enrichment of secondary uranium like in molluscan shells.

There appears to be two ways that fossil corals give uncertain ^{230}Th and ^{231}Pa dates. The one is presence of the "initial ^{230}Th and ^{231}Pa", which were uptaken into the hard tissues together with uranium during fossils' lifetime. The premise to produce ^{230}Th and ^{231}Pa coral ages is the sample in which these daugh-

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ters are initially free. From fossil sample with the initial daughters, therefore, the precise death time of the coral cannot be estimated without a reasonable correction. Any uncorrected dates are regarded as the maximum ages.

Secondly, if the initial thorium and protactinium are distributed in no relation with uranium like at the two structural layers of molluscan shells (OMURA *et al.*, 1973), the heterogeneities in uranium distribution can complicate the picture to secure the reliable dating.

The purpose of this study is to make a close investigation on the amount of thorium and protactinium isotopes and the uranium distribution in present-day hermatypic corals as a basis to understand the extent of potential uncertainty of the observed ^{230}Th and ^{231}Pa dates from fossil corals. The analytical result of the present-day samples can be a direct estimate the undecayed amount of the initial daughters in fossil counterparts grown at the same geographic location, and also can provide the method to correct the apparent ^{230}Th and ^{231}Pa coral dates. If the uranium concentration in a corallum systematically changes, furthermore, the intraskeletal variation of the apparent age obtained may be critically evaluated.

Materials studied

All samples listed in Table 1 are hermatypic corals accompanied by symbiotic zooxanthellae, and most of them were collected alive in order to eliminate any possible postmortem alteration. They were collected from nine locations which extended from Shikoku (Southwest Japan) to New Britain Island along the western part of the Pacific Ocean. These samples are designated CAM-1, CYM-1, CTM-1, CNM-1 and so on, where "C", indicates "coral"; "A", "Amami-o-shima"; "Y", "Yoron-jima"; "T" "Taiwan (Formosa)"; "N", "New Britain Island"; and "M", "modern (present-day)", respectively.

The sea-water samples are coastal water collected from the locations without any distinct influx of land drainage. These samples, however, may not necessarily represent the life-long environment in which the sampled corals grew, because of the possible seasonal variation in local mixing. WY-1 and WY-2 were collected at Chichibina-banare (128°24.2′E, 27°02.8′N) and south of Akazaki (128° 27.3′E, 27°01.1′N), Yoron-jima, respectively; WTk-1, at Hedono (128°53.5′E, 27°48.4′N), Tokuno-shima.

Experiments

Sample preparation Due to low concentration in sea-water, the analysis of uranium and thorium isotopes requires a large amount of sample water; 20*l* of sample water was used for the analysis of uranium and 360*l* for thorium, respectively. After filtered through a cotton gauze in field, suspensions coarser than 0.54 μ in diameter were excluded with a millipore filter.

Carbonate samples for isotopic analysis by α-spectrometry were prepared by the same manner as OMURA *et al.* (1973).

The sample for the fission track analysis was prepared separately depending upon the growth form of the corallum colony, whether branching or massive.

The massive colonial sample (CEM-1) was first embedded in an epoxy resin under vacuum. After solidifying of resin, the sample was trimmed to specific orientation and smoothened the surface by carborundum and chromium oxide powder on polished wheel. And then, it was given similar treatment as tridacnid

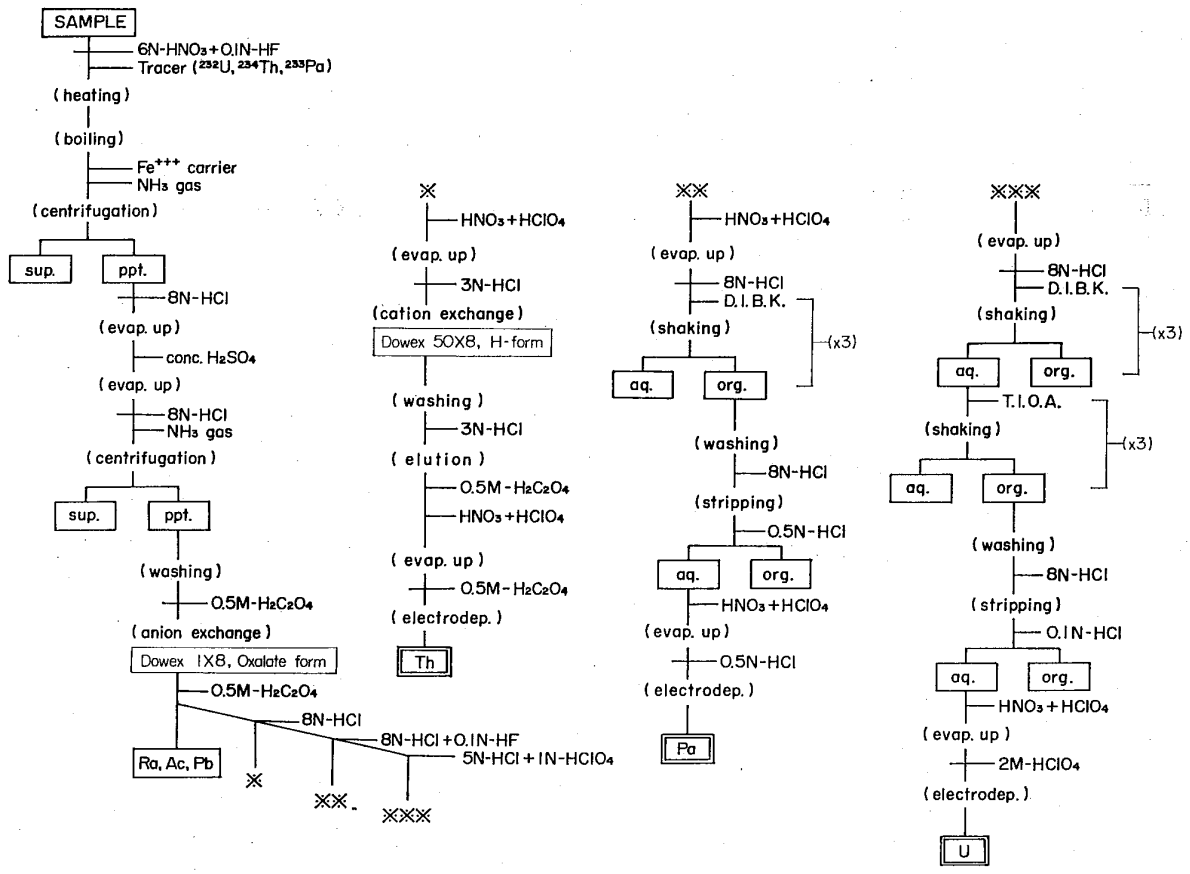
Table 1. List of the present-day coral samples.

Sample	Genus	Location	Collection date
CSM-1*	*Leptoria*	Drift on beach, Muroto-misaki, Shikoku	May 29, 1969
CSM-2*	*Favia*		
CCM-1**	*Favia*	Kominato, Chichi-jima, Ogasawara Islands	Jul., 1969
CCM-2**	*Acropora*	Drift on beach, Okumura, Chichi-jima	Jul., 1969
CHM-1**	*Pocillopora*	Oki-ko, Haha-jima, Ogasawara Islands	Jul., 1969
CAM-1**	*Goniastrea*	Dead sample, Saneku, Amami-o-shima, Ryukyu Islands	Aug., 1968
CEM-1	*Favia*	Kunigami-misaki, Okierabu-jima, Ryukyu Islands	Mar. 28, 1968
CEM-2	*Porites*		
CEM-3	*Favites*	Wanjo-hama, Okierabu-jima	Mar. 27, 1968
CEM-4	*Acropora*	Okidomari, Okierabu-jima	Mar. 24, 1968
CEM-5	*Goniastrea*	Wadomari, Okierabu-jima	Aug. 23, 1967
CEM-6	*Psammocora*	Shira-hama, Okierabu-jima	Aug. 24, 1967
CEM-7	*Acropora*		
CEM-8	*Acropora*		
CEM-9	*Stylophora*		
CYM-1	*Acropora*	N. of Chabana, Yoron-jima, Ryukyu Islands	Mar. 31, 1968
CYM-2	*Acropora*		
CYM-3	*Goniastrea*		
CYM-4	*Acropora*	E. of Aka-zaki, Yoron-jima	Apr. 1, 1969
CYM-5	*Montipora*		
CYM-6	*Porites*		
CYM-7	*Goniastrea*		
CYM-8	*Acropora*		
COM-1*	*Hydnophora*	SW. coast of Kudaka-jima, Ryukyu Islands	Sept. 4, 1960
CTM-1***	*Turbinaria*	Suoa, Taiwan (Formosa)	Date unknown
CNM-1*	*Acropora*	Rabaul, New Britain Island	Feb. 26, 1971
CNM-2*	*Acropora*		
CNM-3*	*Pocillopora*		

* collected by Dr. Kenji KONISHI, Depart. Earth Sci., Fac. Sci., Kanazawa Univ.
** collected by Dr. Yasehide IWASAKI, Geological Institute, University of Tokyo.
*** collected by Dr. T. Y. H. MA of National Taiwan University.

sample (OMURA *et al.*, 1973).

The branching colony as CYM-8 sample was ground carefully with a file, after cleaning throughly. The powdered sample prepared was homogenized for grain size, with sieving between 200 to 250 mesh (74 to 62 μ) screens. The sample was uniformly poured on the muscovite detector placed at the bottom of the polyethylene tube. Both the sample pow-

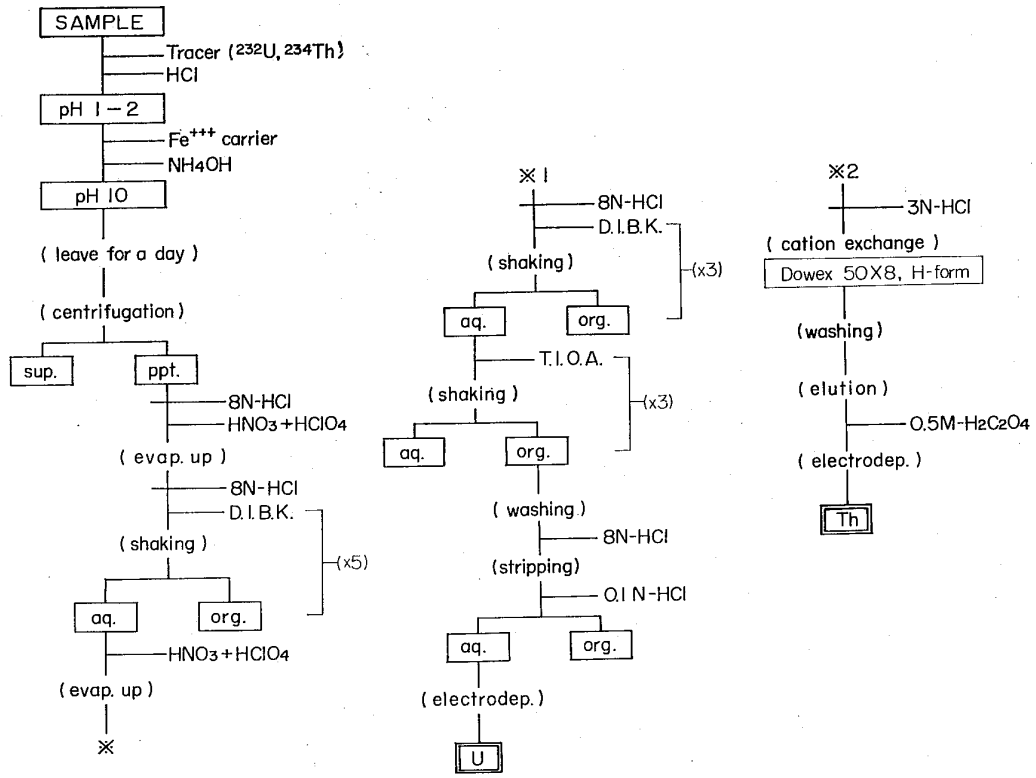


Text-fig. 1. Analytical scheme of the carbonate samples.

der and muscovite detector were fixed with absorbent cotton and a lid of the tube. The reference powder sample with known concentration of uranium was used as the monitor to examine various factors when a uranium concentration was calculated from track density.

α-spectrometry analysis α-spectrometry method was used the isotopic analysis of uranium, thorium and protactinium in carbonate samples and uranium and thorium in sea-water samples. In the present work, it was essentially important to separate each nuclide because of similarity in the energies of the alpha particles emitted by ^{230}Th (4.62 MeV, 4.68 MeV) and by ^{234}U (4.72 MeV, 4.77 MeV). The anion exchange technique established by SAKANOUE *et al.* (KOMURA and SAKANOUE, 1967; SAKANOUE *et al.*, 1967) and partly modified by the author was successful for the separation of each nuclide. The electrodeposition technique was used to prepare the thin sources, which was also important to get a good peak resolution. The separation and purification procedures of each nuclide are schematically summarized in Text-figs. 1 and 2, in the case of carbonate and sea-water samples, respectively. For details of the actual procedures the papers by SAKANOUE *et al.* should be referred to.

Fission track analysis Only the procedures used in this study are briefly outlined below, as the principles of fission track analysis had been detailed already (*e.g.* LAHOUD *et al.*, 1966; SCHROEDER



Text-fig. 2. Analytical scheme of the sea-water samples.

et al., 1970).

Neutron irradiations were carried out in Pn-3 tube of the KUR reactor of Research Reactor Institute, Kyoto University, whose thermal neutron flux density was 2.34×10^{13} n/cm^2•sec. The total neutron flux was determined by the uranium concentration in each sample, which resulted in track density in the order of 10^4 to 10^6 tracks/cm^2.

After etching by 46% hydrofluoric acid solution for 30 minutes at room temperature, fission tracks in muscovite were observed under microscope and photographed. The dimension of the photographed area was calculated from the photograph of the objective micrometer scale with minimum division of 10 μ. The counting areas were placed at random for the powdered sample, but for the solid sample they were located successively along a traverse to map the uranium distribution. The fission tracks on a microphotograph were counted by hand-counting which proved to be fairly rapid and reliable; for example 1,000 tracks in a photograph could be counted in approximately ten minutes.

Results and discussions

The results on isotopic analysis of present-day corals are summarized in Table 2. The standard errors attached to these and other results are derived from counting statistics and does not include any systematic errors.

^{238}U concentration ranges from 2 to 3.5 ppm within the limits of the error. These data agree with those reported previously by many workers (*e.g.* TATSUMOTO and GOLDBERG, 1959; BROECKER and THURBER, 1965; VEEH, 1966; OSMOND *et al.*, 1967; KU, 1968).

Table 2. Analytical results of

Sample	Isotope content			
	^{238}U (ppm)	^{232}Th (ppm)	^{230}Th (dmg)	^{231}Pa (dmg)
CSM-1	2.45±0.10	0.120±0.005	0.0609±0.0020	<0.001
CSM-2	2.65±0.05	0.110±0.005	0.0521±0.0017	0.00501±0.00019
CCM-1	2.75±0.06	0.0511±0.0024	0.0214±0.0009	0.00409±0.00017
CCM-2	3.44±0.07	0.0632±0.0056	0.0249±0.0016	0.00198±0.0008
CHM-1	2.46±0.05	0.0609±0.0040	0.0242±0.0013	0.0140±0.0005
CAM-1	2.22±0.05	0.0275±0.0009	0.0229±0.0005	0.00908±0.00040
CEM-1	2.70±0.07	0.116±0.005	0.0788±0.0022	0.0168±0.0008
CEM-2	2.70±0.07	0.183±0.008	0.0716±0.0033	0.00858±0.00038
CEM-3	2.63±0.05	0.0903±0.0049	0.0517±0.0019	0.00594±0.00026
CEM-4	3.43±0.08	0.0321±0.0018	0.0187±0.0008	0.0113±0.0005
CEM-5	1.90±0.08	0.0517±0.0024	0.0187±0.0007	0.00809±0.00047
CEM-6	2.61±0.13	0.0323±0.0022	0.0215±0.0009	0.0143±0.0005
CEM-7	2.86±0.13	0.0675±0.0054	0.0270±0.0018	0.00700±0.00029
CEM-8	2.75±0.03	0.0386±0.0025	0.0131±0.0007	0.00287±0.00026
CEM-9	2.29±0.06	0.0149±0.0013	0.0149±0.0008	0.00460±0.00017
CYM-1	2.75±0.06	0.0678±0.0033	0.0300±0.0014	0.00402±0.00017
CYM-2a	2.82±0.14	0.0372±0.0021	0.0156±0.0007	0.00343±0.00015
CYM-2b	3.61±0.10	0.0768±0.0040	0.0288±0.0012	0.00421±0.00024
CYM-3	3.51±0.07	0.0442±0.0027	0.0196±0.0009	<0.001
CYM-4	2.99±0.10	0.177±0.009	0.0723±0.0031	0.00142±0.00009
CYM-5	3.02±0.06	0.0337±0.0021	0.0165±0.0007	0.00230±0.00013
CYM-6	2.95±0.07	0.0608±0.0029	0.0224±0.0010	0.00141±0.00009
CYM-7a	2.10±0.04	0.0143±0.0012	0.00700±0.00043	0.0108±0.0005
CYM-7b	2.10±0.03	0.0119±0.0009	0.00550±0.00036	0.0111±0.0005
CYM-7c	2.07±0.04	0.00960±0.00068	0.00451±0.00025	0.00850±0.00028
CYM-7d	2.18±0.03	0.0101±0.0006	0.00384±0.00017	0.00810±0.00031
CYM-7e	2.10±0.02	0.0102±0.0006	0.00486±0.00023	0.00820±0.00030
CYM-7f	1.98±0.03	0.00882±0.00055	0.00412±0.00018	0.00730±0.00030
COM-1	2.67±0.05	0.371±0.019	0.121±0.005	0.0238±0.0015
CTM-1	2.57±0.05	0.0432±0.0041	0.0235±0.0016	<0.001
CNM-1	2.72±0.08	0.109±0.006	0.0360±0.0016	<0.001
CNM-2	3.17±0.14	0.0387±0.0013	0.0125±0.0004	<0.001
CNM-3	3.32±0.08	0.0624±0.0037	0.0215±0.0013	<0.001

the present-day coral samples.

Activity ratio				Apparent age	
$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{230}Th/^{234}U$	$^{231}Pa/^{235}U$	^{230}Th Age	^{231}Pa Age
1.14±0.06	2.13±0.11	0.0296±0.0015	——	3,200±200	——
1.07±0.03	1.98±0.11	0.0251±0.0010	0.0560±0.0023	2,800±100	2,700±100
1.13±0.04	1.76±0.11	0.00938±0.00040	0.0440±0.0021	1,000±50	2,100±100
1.13±0.03	1.66±0.18	0.00873±0.00060	0.0170±0.0007	1,000±70	810±30
1.19±0.03	1.67±0.14	0.0113±0.0007	0.142±0.006	1,200±70	7,200±300
1.13±0.03	3.51±0.14	0.0124±0.0004	0.121±0.006	1,400±40	6,100±300
1.15±0.03	2.86±0.14	0.0346±0.0012	0.184±0.009	3,800±100	9,500±500
1.15±0.03	1.65±0.10	0.0313±0.0013	0.0941±0.0045	3,400±100	4,700±200
1.18±0.03	2.40±0.16	0.0227±0.0009	0.0669±0.0031	2,300±100	3,300±200
1.09±0.03	2.45±0.17	0.00680±0.00032	0.0973±0.0046	760±40	4,800±200
1.18±0.07	1.52±0.09	0.0113±0.0006	0.126±0.010	1,200±60	6,300±500
1.16±0.07	2.80±0.23	0.0112±0.0007	0.162±0.009	1,200±80	8,300±500
1.15±0.07	1.68±0.17	0.0128±0.0010	0.0723±0.0043	1,400±100	3,500±200
1.16±0.03	1.42±0.12	0.00556±0.00030	0.175±0.016	650±40	8,500±800
1.04±0.03	5.34±0.57	0.00850±0.00054	0.0593±0.0026	870±50	2,900±100
1.16±0.03	1.86±0.14	0.0128±0.0006	0.0433±0.0020	1,400±70	2,100±100
1.18±0.08	1.75±0.12	0.00754±0.00048	0.0359±0.0024	870±60	1,700±100
1.10±0.05	1.58±0.11	0.00988±0.00050	0.0345±0.0022	1,100±60	1,700±100
1.09±0.03	1.86±0.14	0.0070±0.0004	——	760±40	——
1.07±0.05	1.72±0.12	0.0307±0.0017	0.0141±0.0010	3,400±200	700±50
1.11±0.03	2.06±0.16	0.00673±0.00032	0.0225±0.0014	740±30	1,100±100
1.08±0.03	1.55±0.10	0.00961±0.00045	0.0141±0.0010	1,100±100	700±50
1.10±0.03	2.06±0.21	0.00412±0.00026	0.153±0.007	450±30	7,800±400
1.10±0.02	1.94±0.20	0.00324±0.00021	0.156±0.008	360±20	8,000±400
1.04±0.03	1.97±0.18	0.00286±0.00017	0.121±0.005	320±20	6,100±300
1.05±0.02	1.60±0.12	0.00229±0.00011	0.110±0.005	250±10	5,500±200
1.06±0.02	2.00±0.15	0.00298±0.00014	0.116±0.005	320±20	5,800±200
1.10±0.02	1.96±0.15	0.00257±0.00012	0.109±0.005	280±10	5,400±200
1.10±0.03	1.37±0.09	0.0563±0.0027	0.264±0.017	5,700±300	14,400±900
1.16±0.03	2.29±0.27	0.0107±0.0007	——	1,200±80	——
1.07±0.04	1.39±0.10	0.0167±0.0009	——	1,800±100	——
1.12±0.07	1.36±0.06	0.00480±0.00026	——	520±30	——
1.13±0.04	1.45±0.13	0.00779±0.00050	——	860±60	——

$^{234}U/^{238}U$ activity ratio in all samples ranges from several to less than twenty percent, which is close enough to the reported value of 1.15 from the present-day corals and oolites by THURBER (1962) and VEEH (1966). This anomaly in the uranium ratio prompted the possibility of an independent dating method, named either "uranium ratio method" or "^{234}U method", which is based on the disintegration of the initial excess ^{234}U in the sample. This method could be useful to cross-check the deficient ^{230}Th and ^{231}Pa dates, but any ages by this method appears to be less reliable. The measurement of $^{234}U/^{238}U$ ratio is necessarily accompanied by the relatively large error as suggested from the gentle slope of the ^{234}U decay curve, and the initial ratio ($^{234}U/^{238}U$ activity ratio in the present-day samples) varies considerably among the individuals even in the same genera at the same habitat. Furthermore, even $^{234}U/^{238}U$ ratio of three sea-water samples in the nearby region is not necessarily constant, though the $^{234}U/^{238}U$ ratio is not unity and show that the activity of ^{234}U is ten or more higher than that of ^{238}U (Table 3).

It might be argumental if the concentrations of thorium and protactinium isotopes in the present-day samples are too low to be determined accurately because of the analytical method used here, but they can hardly be attributed to the analytical contamination as confirmed by repeated blank tests, and can be accepted reliable (Table 2).

In addition to uranium isotopes, measurable amounts of ^{230}Th and ^{231}Pa were detected together with ^{232}Th in the most samples examined. It is clear that corals studied here have initially uptaken thorium and protactinium isotopes into their hard tissues from sea-water. In other words, most of the samples which must show zero year actually produce the initial ^{230}Th and ^{231}Pa ages due to the presence of the initial thorium and protactinium isotopes. These observations probably reflect that the concentrations of thorium and protactinium isotopes are relatively high in sea-water at the sites where the present-day corals examined were collected. The analytical results of sea-water samples support at least the appreciably high concentration of thorium isotopes (Table 3).

Among the samples, the ^{232}Th concentration ranges from 1.7 to 6.0×10^{-9} g/*l*, and the ^{230}Th from 1.3 to 4.0×10^{-14} g/l. Comparing these data with those reported by the previous workers (*e. g.* MOORE and SACKETT, 1964), the observed concentrations of both ^{232}Th and ^{230}Th are taken as relatively higher level within the reasonable limit.

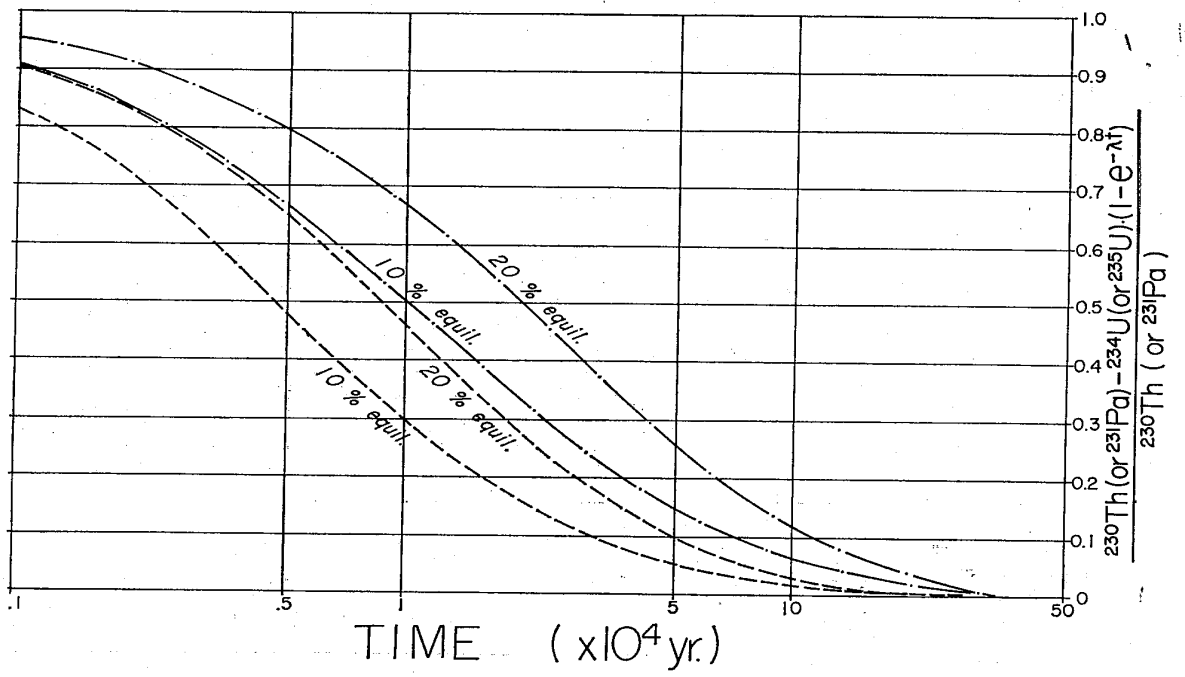
These important observation, coupled with that of the concentration of ^{238}U (2 to 3.5 ppm) and thorium and protactinium isotopes (measurable amounts of both ^{230}Th and ^{231}Pa) in the present-day sam-

Table 3. Summary of α-spectrometry analysis of the sea-water samples.

Sample	Isotope content			Activity ratio	
	^{238}U (10^{-6} g/l)	^{232}Th (10^{-9} g/l)	^{230}Th (10^{-14} g/l)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$
WTk-1	3.76±0.17	5.75±0.27	3.78±0.16	1.15±0.07	1.33±0.09
WY-1	3.54±0.21	5.47±0.25	3.41±0.14	1.11±0.09	1.26±0.08
WY-2	4.31±0.16	1.67±0.15	1.29±0.09	1.16±0.06	1.57±0.18

ples examined, are suggestive of complicated conditions that render both the ^{230}Th and ^{231}Pa dating methods to apply to the fossil corals in the same region. The uncertainty associated with the "apparent ages" due to the initial ^{230}Th and ^{231}Pa, however, is so small that it can be masked with the statistical error and become negligible, in the case of fossils several tens of thousand years old. Within the order of several thousands years, on the contrary, any observed apparent ages may become a matter of serious consideration as discussed below.

Text-fig. 3 was prepared to visualized the effect of the initial ^{230}Th and ^{231}Pa upon the reliability of the apparent ages. Both the initial ^{230}Th and ^{231}Pa decrease with time, while the decay of uranium isotopes is to result in the accumulation of the radiogenic ^{230}Th and ^{231}Pa, in turn. As a whole, both ^{230}Th and ^{231}Pa are expected to increase with time. Text-fig. 3 shows the change of the ratio between the initial and total ^{230}Th and ^{231}Pa with time. The values, 10% and 20% equil. are assigned not only as a matter of convenience, but 10% equil. for ^{230}Th and 20% equil. for ^{231}Pa may be regarded as the maximum values observed in the present-day corals of the Ryukyu Islands. If the initial amount of ^{230}Th and ^{231}Pa were 20% equil., these nuclides would reach more than one-third of the total amounts after 10,000 years, but decrease significantly to approximately several percents beyond 100,000 years. In practice, however, only a few samples exceed 10% equil. in the total amounts of both ^{230}Th and ^{231}Pa. In terms of percent equilibrium with respect to the parent nuclide, ^{231}Pa seems to be contained more abundantly than ^{230}Th in the present-day samples. Thus, the initial ^{231}Pa appears to be more effective to deviate the ap-

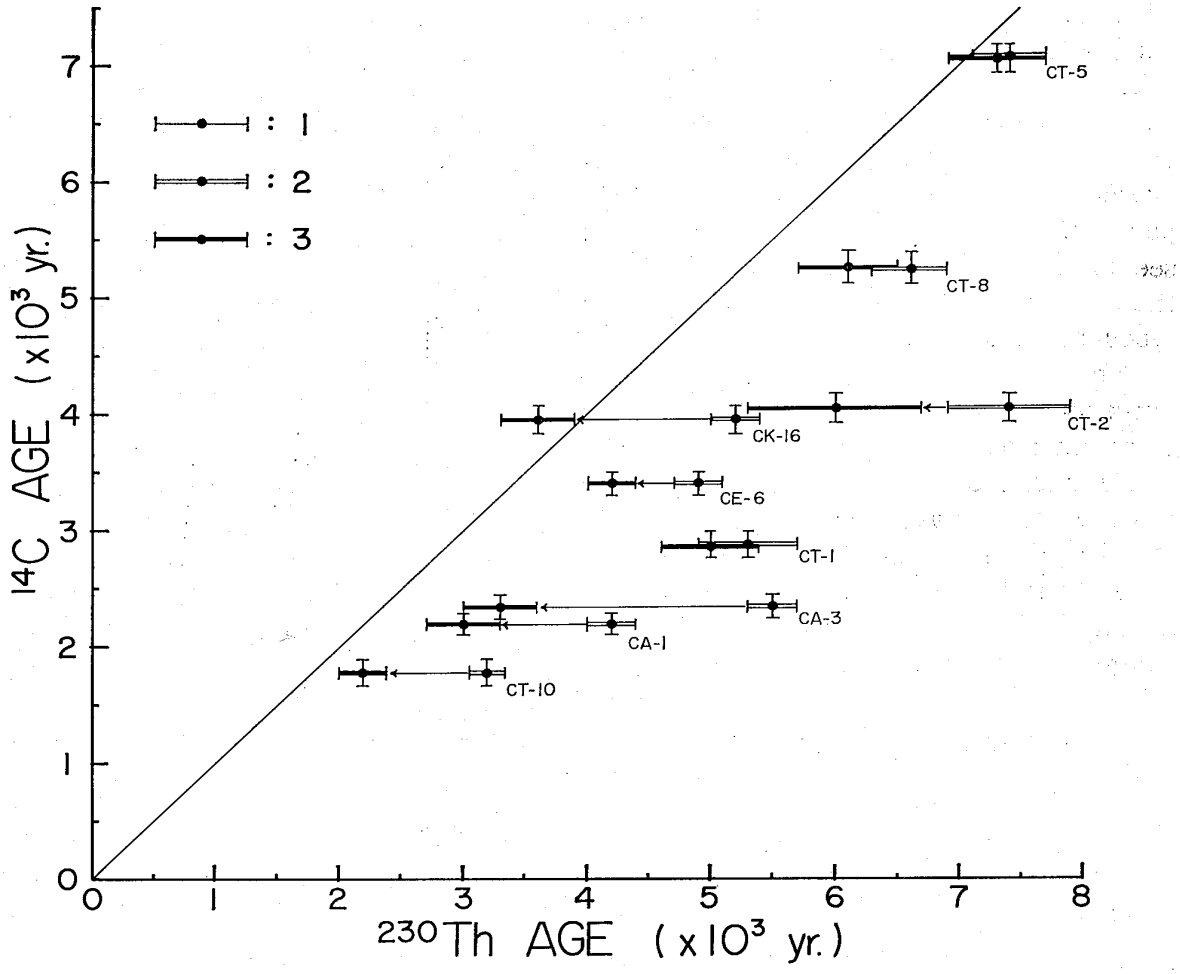


Text-fig. 3. Changes of the initial to total ^{230}Th and ^{231}Pa ratios with time. (The dashed and dotted curves represent the changes of the initial to total ^{230}Th and ^{231}Pa ratio, respectively.)

Table 4. Isotopic composition, apparent and corrected ^{230}Th ages and conventional ^{14}C age of some fossil coral samples.

Sample	Isotope content			Activity ratio			^{230}Th age		^{14}C age* (yr.)	Riken radiocarbon code*
	^{238}U (ppm)	^{232}Th (ppm)	^{230}Th (dmg)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{230}Th/^{234}U$	Apparent age	Corrected age		
CA-1	2.43 ±0.14	0.0856 ±0.0045	0.0807 ±0.0022	1.10 ±0.08	3.97 ±0.23	0.0434 ±0.0024	4,200 ±200	3,000 ±300	2,190 ±120	N 546
CA-3	2.61 ±0.07	0.130 ±0.007	0.107 ±0.003	1.11 ±0.04	3.44 ±0.21	0.0502 ±0.0020	5,500 ±200	3,400 ±300	2,340 ±115	N 548
CK-16	3.28 ±0.11	0.118 ±0.008	0.124 ±0.004	1.11 ±0.05	4.42 ±0.34	0.0463 ±0.0022	5,200 ±200	3,600 ±300	3,950 ±125	N 549
CE-6	2.08 ±0.04	0.0416 ±0.0025	0.0802 ±0.0019	1.16 ±0.03	8.11 ±0.50	0.0454 ±0.0015	4,900 ±200	4,200 ±200	3,410 ±125	N 551
CT-1	2.48 ±0.09	0.0935 ±0.0056	0.0402 ±0.0040	1.11 ±0.06	9.76 ±0.98	0.0493 ±0.0034	5,300 ±400	5,000 ±400	2,880 ±120	N 407
CT-2	2.45 ±0.04	0.0625 ±0.0088	0.127 ±0.010	1.11 ±0.03	8.59 ±0.14	0.0639 ±0.0053	7,400 ±500	6,000 ±700	4,040 ±120	N 399
CT-5	2.57 ±0.09	0.0064 ±0.0005	0.145 ±0.004	1.12 ±0.06	94.8 ±7.5	0.0694 ±0.0033	7,400 ±300	7,300 ±400	7,050 ±145	N 402
CT-8	2.65 ±0.06	0.0275 ±0.0030	0.0935 ±0.0056	1.12 ±0.06	19.4 ±2.1	0.0590 ±0.0030	6,600 ±300	6,100 ±400	5,260 ±140	N 405
CT-10	2.89 ±0.05	0.0488 ±0.0020	0.0713 ±0.0047	1.16 ±0.03	6.15 ±0.48	0.0290 ±0.0020	3,200 ±150	2,200 ±200	1,780 ±115	N 447

* Radiocarbon dates measured by Dr. and Mrs. T. HAMADA at Riken; they are "conventional ^{14}C ages" based on half-life of 5,568 years.



Text-fig. 4. Comparison of the conventional ^{14}C with the apparent and corrected ^{230}Th ages of some fossil corals.

(1, conventional ^{14}C age; 2, apparent ^{230}Th age; 3, corrected ^{230}Th age.)

parent age from the true age than the initial ^{230}Th. But such a deviation in the apparent age will be relatively smaller in the ^{231}Pa age than in the ^{230}Th age for the same fossil sample, because of the shorter half-life of ^{231}Pa.

It is always desirable, if not necessary, evaluate the reliability of a given radiometric date in comparison with that of an independent different radioactive series. Both apparent ^{230}Th and ^{231}Pa ages younger than 35,000 years B. P. can be cross-checked with the ^{14}C ages. Although it is open to question whether the conventional ^{14}C ages represent true ages or not, the discrepancy between the ^{14}C ages and the apparent ^{230}Th ages, as seen in Text-fig. 4, may come at least partly from the presence of the initial thorium isotopes. If the apparent ^{230}Th ages are to be regarded as the ambiguous ones, the next question will be how to reconcile the discordant ages with reasonable accuracy for the samples in which the initial ^{230}Th is contained.

The apparent ages in the deficient ^{230}Th and ^{231}Pa datings are the values calculated on the assumption that all of

^{230}Th and ^{231}Pa are originated from radioactive disintegration of each parent. If the remaining amounts of the initial ^{230}Th and ^{231}Pa in a fossil can be estimated, we can evaluate the amounts of the radiogenic ^{230}Th and ^{231}Pa grown from their parents. Though difficult for ^{231}Pa, it seems to be possible for ^{230}Th to estimate the residual amounts of those initially incorporated.

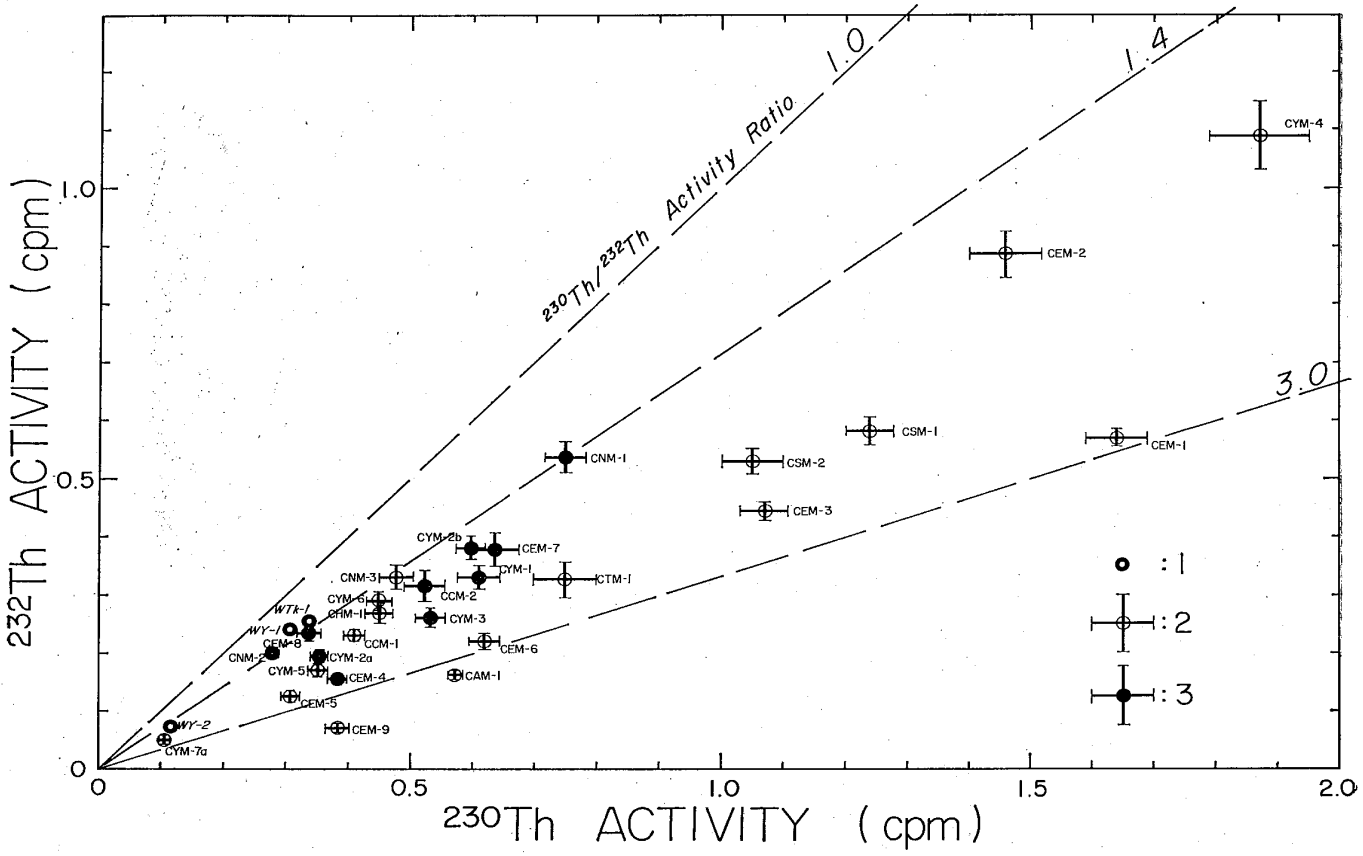
The ^{230}Th/^{232}Th ratios in Ryukyuan present-day corals are characterized by very limited ranges from 1.4 to 3.0 with a few marked exceptions (slightly higher than 2.0, on an average; Text-fig. 5). On the other hand, the decay of ^{232}Th is so slow that the decrease in amount of the initial ^{232}Th can be ignored for the time range of the deficient ^{230}Th dating. If the assumption can be made that the initial ^{230}Th/^{232}Th ratios in fossil remains had been the same degree of constancy as those in their present-day counterparts, the residual initial ^{230}Th could be estimated by the equation from the ^{232}Th concentration, as follows;

$$^{230}\mathrm{Th}_{\mathrm{initial}} = \mathrm{R} \cdot {}^{232}\mathrm{Th} \cdot e^{-\lambda t} \quad \cdots\cdots (1)$$

where λ is the decay constant of ^{230}Th, and R is the initial ^{230}Th/^{232}Th ratio and defined by the following equation;

$$\mathrm{R} = \left(\frac{^{230}\mathrm{Th}}{^{232}\mathrm{Th}}\right)_{t=0} = \frac{^{230}\mathrm{Th} - {}^{234}\mathrm{U} \cdot (1 - e^{-\lambda_{234} t})}{^{232}\mathrm{Th} \cdot e^{-\lambda_{232} t}} \quad \cdots\cdots (2)$$

where λ_{234} and λ_{232} are decay constant of ^{234}U and ^{232}Th, respectively. Then the radiogenic ^{230}Th (^{230}Th$_{\mathrm{rad.}}$) can be calcu-



Text-fig. 5. ^{230}Th/^{232}Th activity ratios in sea-water and present-day coral samples. (1, sea-water samples; 2, massive colonial corals; 3, branching colonial corals.)

lated by the following equation;

$$^{230}Th_{rad.}=^{230}Th_{total}-R\cdot^{232}Th\cdot e^{-\lambda t}\cdots\cdots(3)$$

If the uncertainty of an apparent ^{230}Th age depends only on the amount of the initial ^{230}Th, it can be satisfactorily solved by the proper estimation of the R value. The R values have been previously estimated from either the analytical data of sea-water or comparison between ^{14}C and ^{230}Th dates of fossil molluscs and corals (KAUFMAN and BROECKER, 1973; THURBER *et al.*, 1965; VALLENTINE and VEEH, 1969; VEEH and CHAPPELL, 1970; KAUFMAN *et al.*, 1971). In this study, this value was determined directly by analyses of present-day corals and sea-water where corals were sampled.

The average ^{230}Th/^{232}Th ratio in the examined sea-water samples of the Ryukyu Islands is 1.4 (Table 4), whereas the present-day coral samples have a little higher ratio as described above. In the case of the present-day massive corals such as *Favia* and *Goniastrea*, the analyses were carried out at the inner portion of the corallum away from the surface where polyps were attached, in order to avoid the contamination of nuclides from soft tissue. If the sample was taken about 5 cm inside from the surface of a massive colony, *Favia*, it would represent an growth increment approximately about 10 years old, if a growth rate is accepted to be 0.5 cm/year (OMURA *et al.*, 1972). In this case, all ^{230}Th in the portion analyzed may not be incorporated through secretion process of the skeleton, but might have grown in part from radioactive disintegration of ^{234}U during the last ten years.

Then, the apparent increase of ^{230}Th/^{232}Th ratio in "present-day coral" samples can be explained by assuming the initial isotopic composition as follows:

^{238}U=2.7 ppm

^{234}U/^{238}U=1.15 A. R.

^{232}Th=0.05 ppm.

These values are not absurd assumption but fair average among the present-day corals of the Ryukyu Islands. The results of the calculation are shown in Table 5 which lists data for growth ages versus apparent changes of the R value. After all, it seems acceptable to adopt the ^{230}Th/^{232}Th ratio of the sea-water in habitat regardless of taxa of the organisms, as the R value.

Table 5. Apparent changes of R values (the initial ^{230}Th/^{232}Th ratios) with growth ages.

Age (years old)	0	10	20	30	40	50
R	1.0	1.2	1.4	1.6	1.8	1.9
	1.5	1.5	1.9	2.1	2.3	2.4
	2.0	2.2	2.4	2.6	2.8	2.9

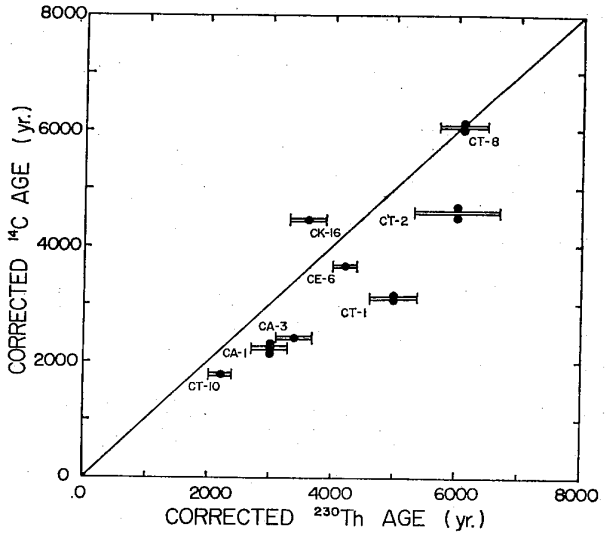
^{231}Pa also is probably incorporated initially along with thorium isotopes. With our present knowledge, however, it is not possible to estimate the residual amounts of the initial ^{231}Pa in fossil remains by any logical steps.

Thus, the corrected apparent ^{230}Th ages, substituting 1.4 as the R value for equation (3), were compared again with the conventional ^{14}C ages (Text-fig. 4). The discrepancies between the corrected ^{230}Th and ^{14}C ages undoubtedly become somewhat smaller, although they are not in entire agreement. These discrepancies between two dates may be attributable to the uncertainty inherent to the R value. However, there exists uncertainty for the conventional ^{14}C ages.

The ^{14}C age is conventionally computed, in principle, assuming that all the samples

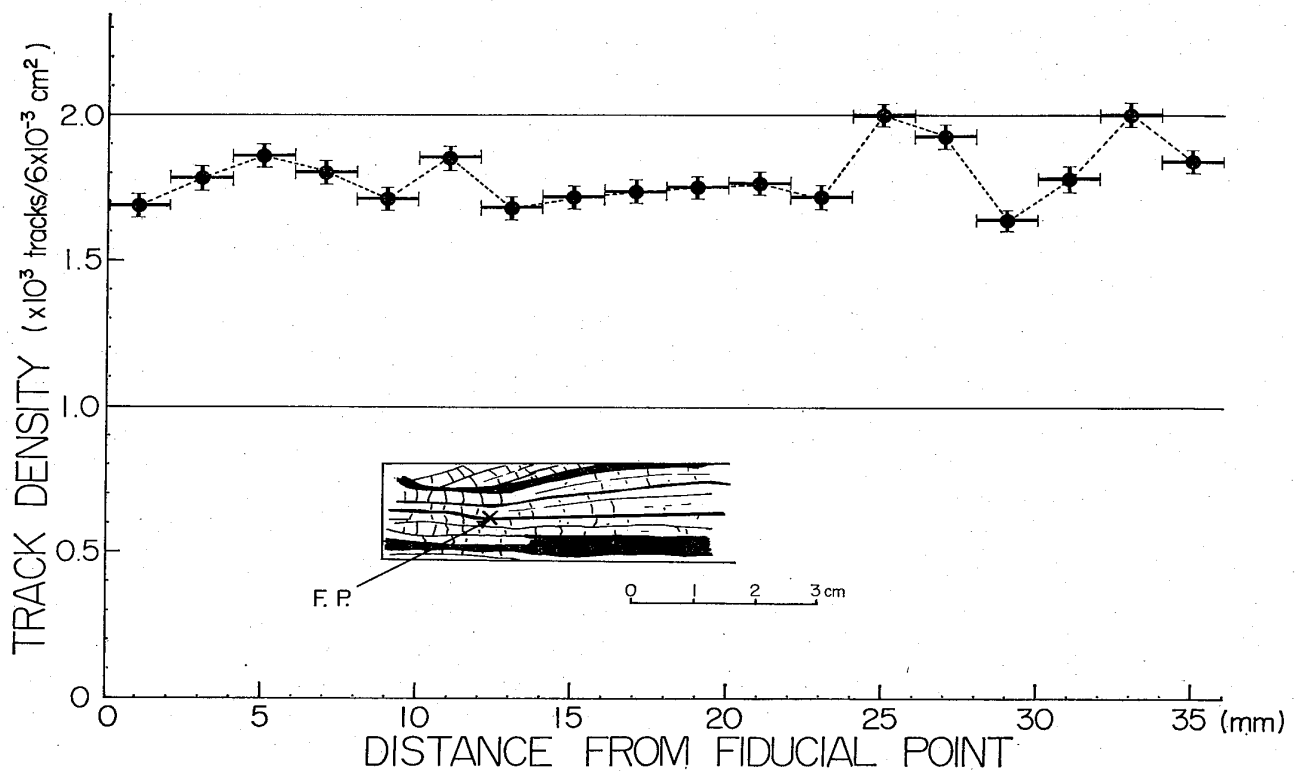
initially contained the same $^{14}C/^{12}C$ ratio. If this assumption was not accepted, the Conventional ^{14}C age may not be relied upon. SUESS (1965, 1967, 1969) and KIGOSHI and HASEGAWA (1966) inferred from the discrepancy between dendrochronometric ages of wood samples and their ^{14}C ages that $^{14}C/^{12}C$ ratio, or ^{14}C content in atmosphere has changed during the past 6,000 years. Additionally they proposed the correction curves for the conventional ^{14}C ages.

Text-fig. 6 compares the corrected ^{230}Th age with the ^{14}C age converted graphically with the correction curve of SUESS (1969). From the text-figure, it seems as if the discrepancies between two corrected dates reduce further smaller. If the correction of ^{230}Th ages proposed here can be substantiated with future works, a similar correction curve of the conventional ^{14}C ages may be obtained beyond the dendrochronometric age, based on the ^{230}Th ages of hermatypic corals.

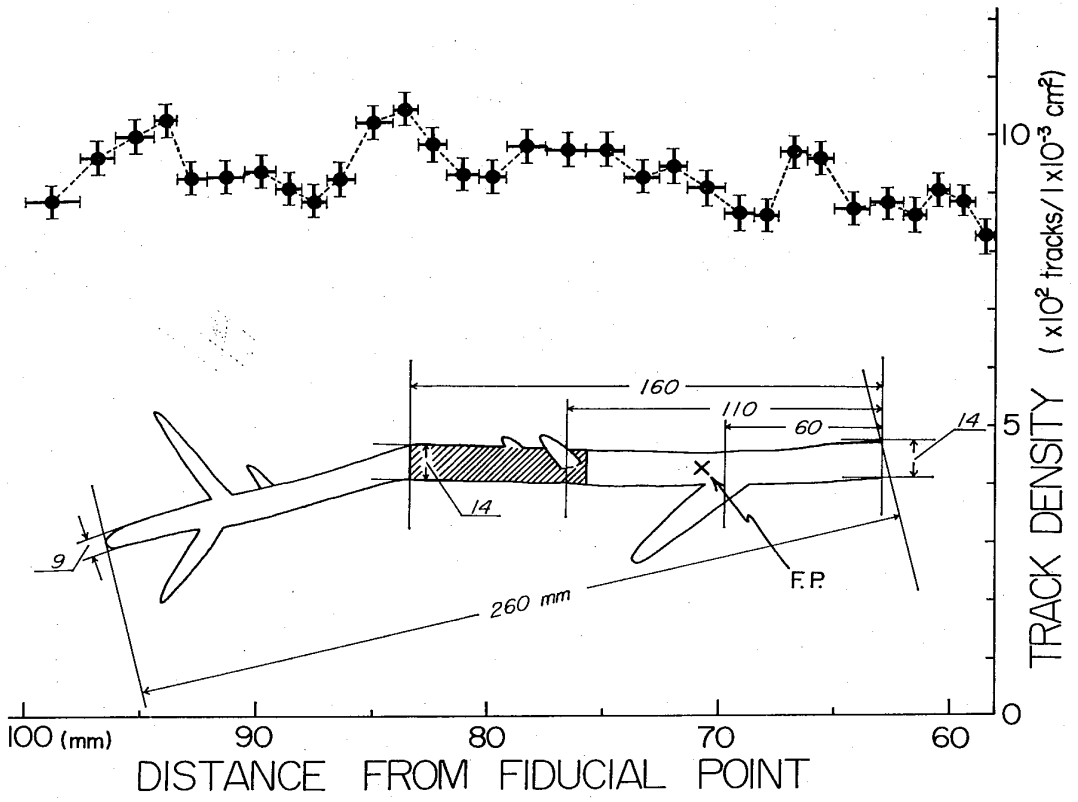


Text-fig. 6. Comparison between corrected ^{230}Th and ^{14}C dates.

The results of the fission track analysis



Text-fig. 7. Fission track counts measured along an axial line on a longitudinal section of CEM-1 specimen.



Text-fig. 8. Variation of track density in the axial growth direction of a branching coral (*Acropora* sp., CYM-8).

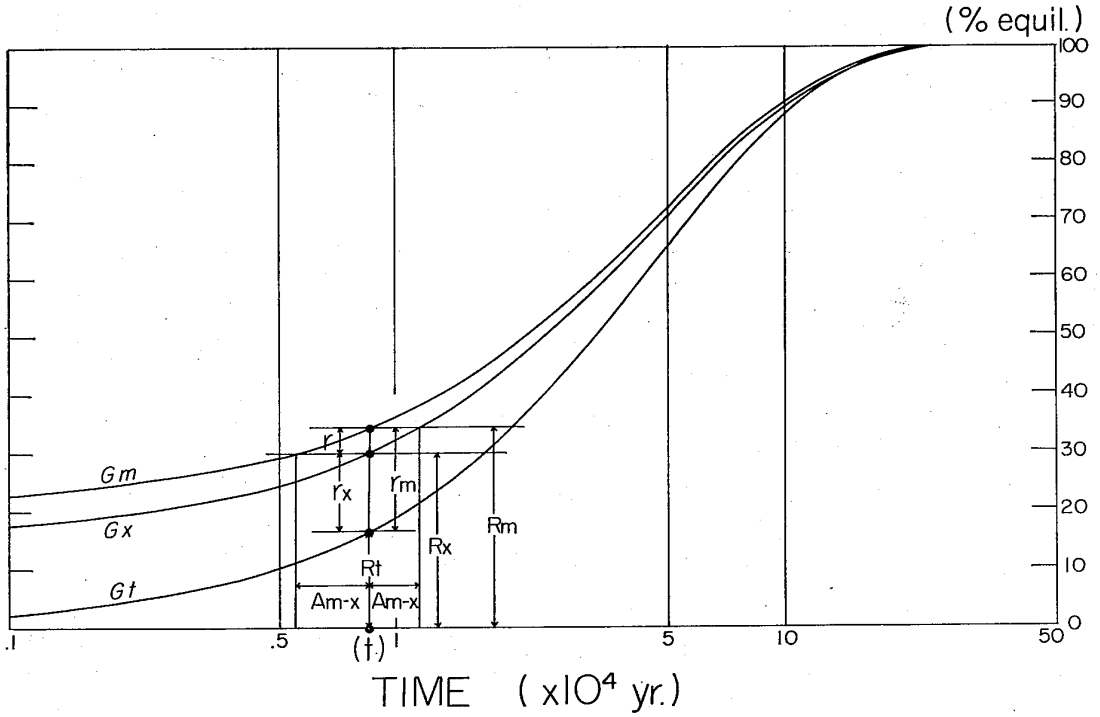
to examine the uranium distribution in coral skeletons are given in Text-figs. 7 and 8 where uranium distribution is expressed as the relative changes of track density.

Text-fig. 7 is the results obtained by counting the fission tracks along an axial line on a first-cycle septum in which no growth increments are recognized morphologically. Text-fig. 8 shows the uranium distribution in axial growth direction of a branching corallum (*Acropora* sp.). From these illustrations, it is clear that uranium distributes not necessarily in uniform but heterogeneously. The variation of the uranium concentration is estimated to be 30% at the maximum, although the much profound differences like those between two structural layers of tridacnid shells are not confirmed (OMURA *et al.*, 1973).

Assumed that both ^{230}Th and ^{231}Pa are distributed homogeneously, in contrast to the heterogeneities of uranium, the difference in isotopic composition can be calculated by combining α-spectrometry and fission track analyses for the three respective parts in which uranium isotopes are maximum, average and minimum in concentration. As to the analytical results of CEM-1 (Table 6), the following possibilities may be pointed out. If this sample had been well preserved as a closed system during its diagenetic history, ^{231}Pa should grow from its parent ^{235}U as shown in Text-fig. 9. The growth curve of ^{230}Th might have the same pattern as that of ^{231}Pa. The theoretical growth curve (Gt) estimated from the sample in which the initial ^{231}Pa is free, also is shown together in text-figure. We notice readily by assuming the arbitary

Table 6. Difference in isotopic composition among the various portions in a present-day coral sample (U_x, the maximum portion in uranium concentration; U_a, the average portion in uranium concentration; U_m, the minimum portion in uranium concentration).

	Activity (dmg)				Activity ratio		Apparent age (yr.)	
	^{238}U	^{234}U	^{230}Th	^{231}Pa	$^{230}Th/^{234}U$	$^{231}Pa/^{235}U$	^{230}Th age	^{231}Pa age
U_x	2.28	2.62	0.788	0.0168	0.0301	0.160	3,300	8,200
U_a	1.98	2.28			0.0346	0.184	3,800	9,500
U_m	1.72	1.98			0.0398	0.212	4,400	11,000



Text-fig. 9. Growth curve of ^{231}Pa estimated in CEM-1 sample.
(G_t, theoretical growth curve; G_m, growth curve estimated in the minimum part in uranium concentration; G_x, growth curve estimated in the maximum part in uranium concentration.)

time (t) that the $^{231}Pa/^{235}U$ ratio of CEM-1 is r_m or r_x higher than that of the ideal sample. Moreover, there should be the difference (r) in $^{231}Pa/^{235}U$ ratio, between the maximum and minimum parts of uranium isotopes in concentration. This r is responsible for the partial difference in apparent ^{231}Pa age, which is represented as A_{m-x} in Text-fig. 9. The decrease of r with time must be noted at this point. The relation between r and time can be seen also in Table 7 which shows the magnitude of A_{m-x} in various stages. Namely, the older the sample, the smaller the uncertainty due to heterogeneities in uranium distribution

Table 7. Changes of A_{m-x} values in various stages (see text for detailed discussion on A_{m-x} value).

Time (yr.)	A_{m-x} (yr.)	
	^{230}Th age	^{231}Pa age
0	1,100	2,800
1,000	1,100	2,800
5,000	1,000	2,700
10,000	1,000	2,400
50,000	700	900
100,000	500	300

appears to be the general rule. After more than several tens of thousand years, this uncertainty in apparent ages of CEM-1 sample may become too small to be distinguished from the statistical error of counting. However, it is undoubtedly one of the unfavorable factors for reliable dating, if a sample is younger than 10,000 years old.

The above discussion may not fit in the reality, because we possess but very incomplete information on thorium and protactinium isotopes distribution in various present-day organisms. If ^{232}Th microanalysis using fission tracks (HAIR *et al.*, 1971) be developed, we may be able to obtain the distribution patterns of ^{232}Th in skeletal carbonates.

Conclusion

The α-spectrometry combined with fission track analysis was performed to get informations as to the concentrations and the activity ratios of uranium, thorium and protactinium isotopes in some present-day and fossil hermatypic corals, and to examine critically the effectiveness of the deficient ^{230}Th and ^{231}Pa dating methods.

The analysis of the sea-water samples from the Ryukyu Islands revealed that uranium was contained to the same extent as the sea-water in the other regions, while the concentrations of thorium isotopes were a little higher than those in the mid-oceanic region. These results vindicate that marine organisms are generally grown under the same environment with respect to uranium, but not necessarily so for thorium isotopes (and probably protactinium isotopes as well).

The ^{230}Th and ^{231}Pa concentrations were not low enough to be neglected in the observed present-day corals. Because of such high ^{230}Th/^{234}U and ^{231}Pa/^{235}U ratios, the present-day coral samples examined had the initial ^{230}Th and ^{231}Pa ages. From tne presence of the equivalent amounts of ^{232}Th, it may be inferred with certainty that the Ryukyuan fossil corals initially withheld similar ^{230}Th/^{234}U ratio.

If it is a safe assumption that the fossils initially had the constant ^{230}Th/^{232}Th ratio similar to the present-day counterparts in the same region, the residual amounts of the initial ^{232}Th can be estimated by the following equation from the ^{232}Th concentration:

$$^{230}\mathrm{Th}_{\mathrm{initial}} = \mathrm{R} \cdot {}^{232}\mathrm{Th} \cdot e^{-\lambda t}$$

where λ is the decay constant of ^{230}Th, and R is the initial ^{230}Th/^{232}Th ratio. The uncertainty of an apparent ^{230}Th age, due to the presence of the initial ^{230}Th, can be satisfactorily solved by the proper estimation of the R value.

The present-day corals apparently had higher ^{230}Th/^{232}Th ratio than the sea-water in their habitat. Accordingly, it could be questioned to adopt the ^{230}Th/^{232}Th ratio of the present-day coral samples as the R value. The higher ^{230}Th/^{232}Th in the present-day samples is accounted for by the growth rate of herma-

typic corals and the concentration of ^{234}U enough to let the initial $^{230}Th/^{232}Th$ ratio grow for several decades. After all, for the Ryukyuan fossil corals, it seems reasonable to use 1.4, an average $^{230}Th/^{232}Th$ ratio of the coastal water in habitat, as R value.

In the Ryukyuan fossil corals in which the initial ^{230}Th and ^{231}Pa are contained, the difference in distribution pattern between uranium isotopes and their daughter nuclides also become a troublesome problem. Although an extreme case comparable to the two structural layers of molluscan shell had not been confirmed, the differences in uranium concentration to attain 30% at the maximum were observed in axial growth direction of two scleractinian corallites. If the initial thorium and protactinium isotopes were distributed in no relation with uranium isotopes, the apparent ^{230}Th and ^{231}Pa ages may be partially different, and the different growth curves of ^{230}Th and ^{231}Pa must be used for dating.

The age ambiguity due to the initial daughters and the heterogeneities in uranium distribution must always be evaluated critically, but these factors may not cause any practical effect on dating the samples older than tens of thousand years.

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