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744. URANIUM-SERIES AGE OF THE "KAMETSU FORMATION", RIUKIU LIMESTONE ON THE TOKUNO-SHIMA, RYUKYU ISLANDS*

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Abstract. Uranium-series dating of some hermatypic corals (*Acropora* sp.) reveals that the uppermost part of the Pleistocene series occurred on Tokuno-shima (Central Ryukyus) is likely to be Middle Pleistocene in age, at least older than 300,000 years. Although the coral ages obtained are almost near or beyond the limitation of the $^{230}Th/^{234}U$ method of dating, the upper limit of the age is estimated from the mean $^{234}U/^{238}U$ activity ratio to be approximately 700,000 years. This result is suggestive that reefy sediments formed during the last interglacial stage, 120,000 eyars ago when sea level stood about 6 m higher than the present, is exposed nowhere on the island. In addition, the absence of the Holocene Raised Coral Reef Limestone (Hanzawa, 1935) on the island may also imply that the vicinity of Tokuno-shima has been locally subsided since the last 120,000 years.

The present paper deals with radiometric ages estimated by the $^{230}Th/^{234}U$ and $^{234}U/^{238}U$ methods for hermatypic corals from the uppermost part of the Pleistocene series on Tokuno-shima, Central Ryukyu Islands. These are the first uranium-series dates from this island, which allow us to correlate the dated stratigraphic unit with those on the other islands in Ryukyus and to relate it to various Pleistocene events in other areas.

Most of all islands in Central and Southern Ryukyus are capped or rimmed with Quaternary reefy limestone which was divided into two stratigraphic units, Pleistocene Riukiu Limestone and Holocene Raised Coral Reef Limestone by Hanazawa (1935). Subsequently, the Riukiu Limestone has been subdivided into a few unit on some islands. For an example, Nakagawa (1967) redefined the Pleistocene series on Tokuno-shima as the Ryukyu Group and stratigraphically subdivided into three, Kametsu, Kinoko and Itokina Formations, in descending order, each of which was morphostratigraphically correlated among subdivisions on the neighboring islands, Okierabu-jima, Yoron-jima and Kikai-jima (Nakagawa, 1969). It presents, however, practical difficulty to determine the age of each unit by means of a radiometric dating such as $^{230}Th/^{234}U$ method, because organic remains of calcium carbonate are diagenetically altered in most cases to stable phase (low Mg calcite) in phreatic and/or vadose environments. Moreover, morphostratigraphic technique also can be hardly employed to correlate among subdivisions on respective islands, because of the local variation in the rate of vertical displacement (Konishi *et al.*,

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1974). Those reasons have made it hard to assign chronologically the formation of Pleistocene series in Ryukyus, even though it was geologically investigated in detail and stratigraphically subdivided into some units.

The coral samples analyzed here are only a genus *Acropora* collected at Isen-zaki (27°39.4′ N Lat., 128°56.3′ E Long.), the southernmost end of Tokuno-shima (Fig. 1), where a narrow (ca. 100 m in width) but cospicuous marine terrace is developed over a distance of several kilometers along the coast. The top of the terrace is approximately 9 m in altitude. As illustrated in Fig. 2, the limestone unit which overlies a basal conglomerate is about 7 m in the maximum thickness. The uppermost part of the limestone unit is abundant in coral heads (mostly of *Acropora* sp.), over 100 cm in diameter in growth position. During five days for an on-the-spot investigation, fourty-eight coral heads were carefully examined for their mineralogical nature by the staining method using Feigl's solution in an area of about 20,000 m² on the surface of the terrace, and fifteen samples were chosen for their appearance of aragonitic nature and took back to the laboratory.

The terrace sediments shown in Fig. 2

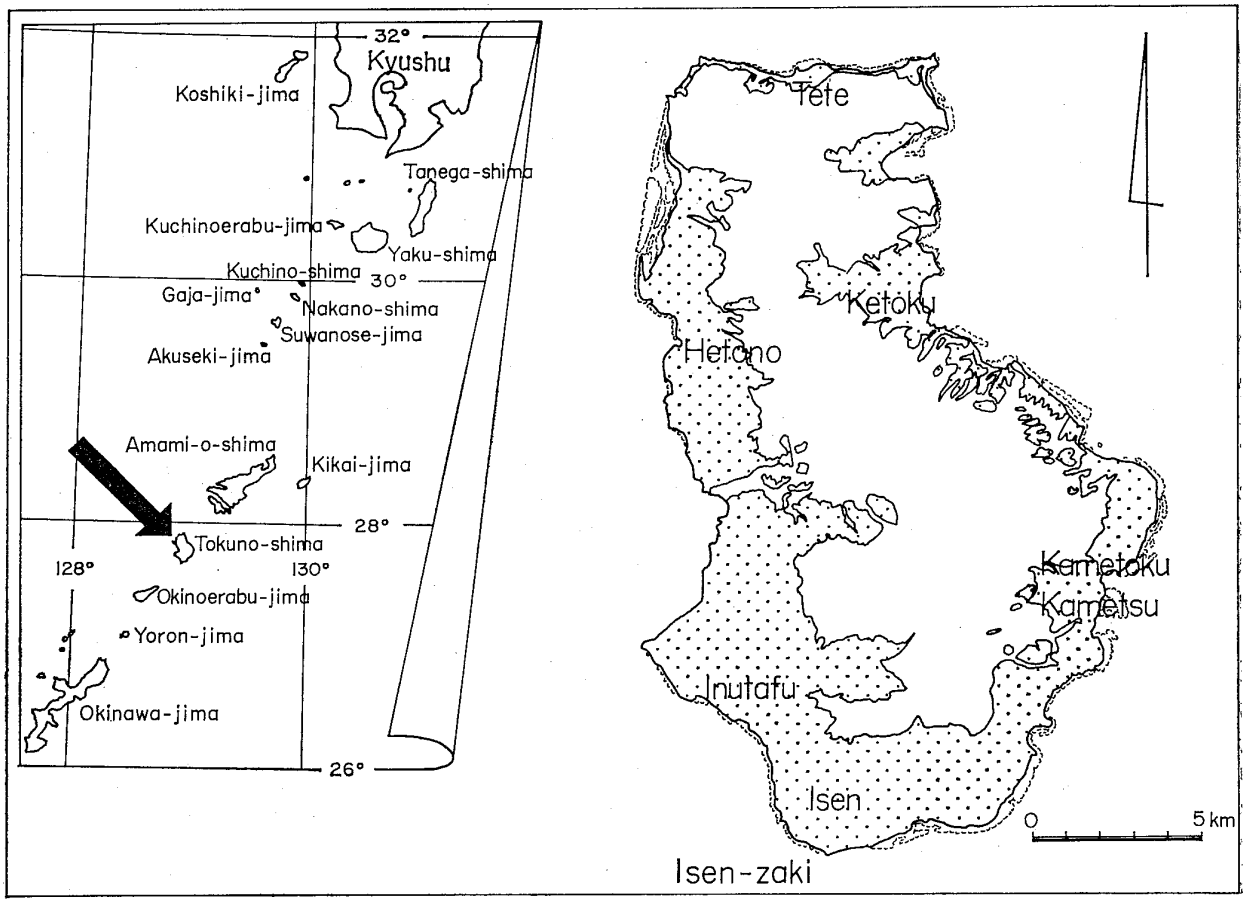

Fig. 1. Index map showing the locality where the fossil coral samples were collected. (Dotted part is the distributing area of Pleistocene series on Tokuno-shima; after Nakagawa, 1967).

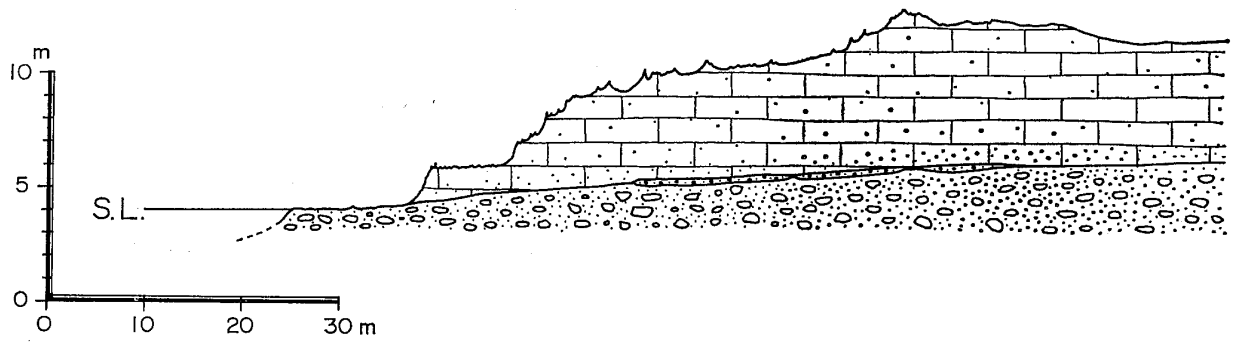

Fig. 2. Simplified sketch of the terrace sediments at Isen-zaki, the southernmost end of Tokuno-shima.

come under a part of the Kametsu Formation of Nakagawa (1967) and are regarded as the uppermost part of the Pleistocene series on Tokuno-shima. The ^{14}C dates of 5,800±230 (Machida *et al.*, 1976) through $43{,}700^{+7{,}000}_{-5{,}800}$ years B. P. (seen in the table compiled by Pirazzoli, 1978) have been reported for some corals which were collected at the same locality as the sampling site in this study. The Research Group for Active Faults (1980) seems to consider that the limestone units formed during two stages (120,000 and less than 100,000 years ago, respectively) are occurred along Isen-zaki.

Results of analyses are summarized in Table 1.

X-ray powder diffraction patterns revealed that low Mg calcite appeared owing to the recrystallization of skeletal aragonite was detectable in most samples. It was only one sample numbered as 80-2-9-5 that was free of the secondary calcite. In the present study, the analyses were repeated for this and the other two samples (Nos. 75-11-10-1 and 75-11-10-2) in which only a few percent or less in weight of calcite was detected. Two sorts of ^{232}U-^{228}Th spikes, namely "Harwell" spike allotted by the Uranium-Series Intercomparison Project (Harmon *et al.*, 1979) and "KU (Kanazawa Univ.)" spike prepared by the author, were used as yield tracers in the repeated analyses, separately. Consequently, the high reproducibility of the results attests to the reliability of each value listed in Table 1. $^{230}Th/^{234}U$ ages were calculated by the equation;

$$^{230}Th = {}^{238}U[1-\exp(-\lambda_0 t)] + ({}^{234}U - {}^{238}U)[\lambda_0/(\lambda_0-\lambda_4)] \cdot [1-\exp(\lambda_4 t-\lambda_0 t)]$$

where λ_0 and λ_4 are decay constants of ^{230}Th and ^{234}U, respectively. The quoted errors are based on the counting statistics.

The samples containing five percent or more of calcite cannot be anticipated showing the reliable uranium-series dates, because a closed system respect to uranium and thorium isotopes has not been retained throughout their diagenetic history. The upper six $^{230}Th/^{234}U$ ages for three samples in Table 1, however, are reliable enough to use in the discussion below for the following evidences; (1) The specimens are entirely or almost free of recrystallization, as shown by the absence or an extremely small quantity of the secondary calcite. (2) The assumption of negligible initial ^{230}Th is supported by the observation that two samples seem to be free of ^{232}Th and that $^{230}Th/^{232}Th$ ratios in the other one

Table 1. Isotopic composition and estimated ages of fossil corals from the Kametsu Formation.

Sample No.	Calcite (wt %)	Isotope Concentraion				Activity Ratio			Estimated ^{230}Th Age (ka)
		^{238}U (ppm)	^{234}U (dpm/g)	^{232}Th (ppm)	^{230}Th (dpm/g)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{230}Th/^{234}U$	
80-2-9-5	0	3.07±0.05	2.30±0.04	< 0.02	2.32±0.04	1.01±0.02		1.01±0.02	> 468
		3.02±0.05	2.33±0.04	< 0.02	2.29±0.04	1.03±0.01		0.983±0.021	$423^{+\infty}_{-78}$
75-11-10-1	< 1	3.52±0.08	2.70±0.06	< 0.02	2.70±0.06	1.03±0.02		1.00±0.03	$486^{+\infty}_{-134}$
		3.41±0.09	2.60±0.07	< 0.02	2.60±0.06	1.02±0.02		1.00±0.03	$529^{+\infty}_{-169}$
75-11-10-2	3	3.34±0.04	2.58±0.03	0.0464±0.0085	2.63±0.06	1.04±0.01	236 ± 43	1.02±0.03	> 407
		3.42±0.05	2.55±0.04	0.0257±0.0034	2.46±0.03	1.02±0.01	392 ± 52	0.943±0.018	301^{+35}_{-27}
80-2-8-3	5	3.22±0.04	2.41±0.03	< 0.02	2.50±0.04	1.00±0.01		1.04±0.02	∞
80-2-9-2	5	3.26±0.04	2.45±0.03	0.0441±0.0077	2.07±0.04	1.01±0.01	196 ± 34	0.845±0.018	201^{+13}_{-12}
80-2-9-4	9	2.88±0.04	2.14±0.03	< 0.02	1.69±0.06	1.00±0.01		0.790±0.028	169^{+16}_{-13}
75-11-10-4	21	2.83±0.07	2.11±0.05	< 0.02	2.00±0.08	1.00±0.02		0.948±0.045	320^{+220}_{-70}
75-11-10-5	43	2.59±0.06	1.93±0.04	0.114±0.016	2.25±0.06	1.00±0.02	82.3±11.8	1.17±0.04	∞

(For the upper three samples, the results in the upper and lower rows were obtained by use of "Harwell" and "KU" spikes as a yield tracer, respectively.)

(75-11-10-2) are very much higher than those in natural waters, which are commonly 1 to 3.

The $^{230}Th/^{234}U$ ratios for the upper three samples in Table 1 range from 0.943 ± 0.018 to 1.02 ± 0.03. These values are suggestive that ^{230}Th has been nearly attained equilibrium with its parent ^{234}U. For this reason, the limestone unit from which the samples examined were derived appears to be too old for the $^{230}Th/^{234}U$ method of dating to be applicable, although the ages can be numerically calculated by using the above equation, as shown in the table. In other words, the Kametsu Formation of Nakagawa (1967) is regarded as being more than 300,000 years old.

Nakagawa (1969) attempted once to correlate morphostratigraphically the Kametsu Formation with his Wan Formation on Kikai-jima. On the other hand, the Pleistocene series on Kikai-jima was subdivided into four units, Araki Limestone, and Younger, Middle and Older Limestone Members of Riukiu Limestone, in descending order by Konishi *et al.* (1974), which have been dated by the $^{230}Th/^{234}U$ and $^{231}Pa/^{235}U$ methods to be 35,000-45,000, 55,000-65,000, 80,000-100,000 and 120,000-130,000 years, respectively. The Wan Formation of Nakagawa (1969) seems to be matchable a suit from the Araki Limestone through the Middle Limestone Member of Riukiu Limestone of Konishi *et al.* (1974). In this viewpoint, it may be said for the above-mentioned date of the Kametsu Formation to be too old contrary to the expectations. However, the reliability of an age of more than 300,000 years is supported by the average value of $^{234}U/^{238}U$ ratios in Table 1.

The $^{234}U/^{238}U$ activity ratio can be used to estimate the age of biogenic carbonates by the following equation, provided that the initial activity ratio (Ro) is known and the sample has remained closed to uranium and the daughters between ^{238}U and ^{234}U in the uranium decay series:

$$^{234}U/^{238}U = 1 + (Ro - 1)e^{-\lambda_{234}t}$$

Because three values for Ro have been reported up to present, the equation is plotted in Fig. 3 to show the decrease of $^{234}U/^{238}U$ ratio as a function of time for the cases, each of which Ro is 1.15 (Thurber, 1962), 1.14 (Ku *et al.*, 1977) and 1.12 (Nikolayev *et al.*, 1979), respectively.

The $^{234}U/^{238}U$ ratios for the three samples are characterized by limited ranges of 1.01 ± 0.02 to 1.04 ± 0.01, and the mean is calculated to be 1.03 ± 0.01. From this mean $^{234}U/^{238}U$ ratio, the age of the Kametsu Formation is estimated to be 387,000 through 709,000 years. Finally, it may be given as a conclusion here that the uppermost part of the Pleistocene series on Tokuno-shima is 300,000 years of age or more, the upper limit of which is estimated as being approximately 700,000 years.

It follows justly from a conclusion stated above that a question arises as to the reefy sediments which must have been formed around Tokuno-shima during high stand of the sea 120,000 years ago. Shoreline features standing about 6 m above the present sea level have been found in many places in the world, and they are thought to represent a eustatic high sea stand that occurred 120,000

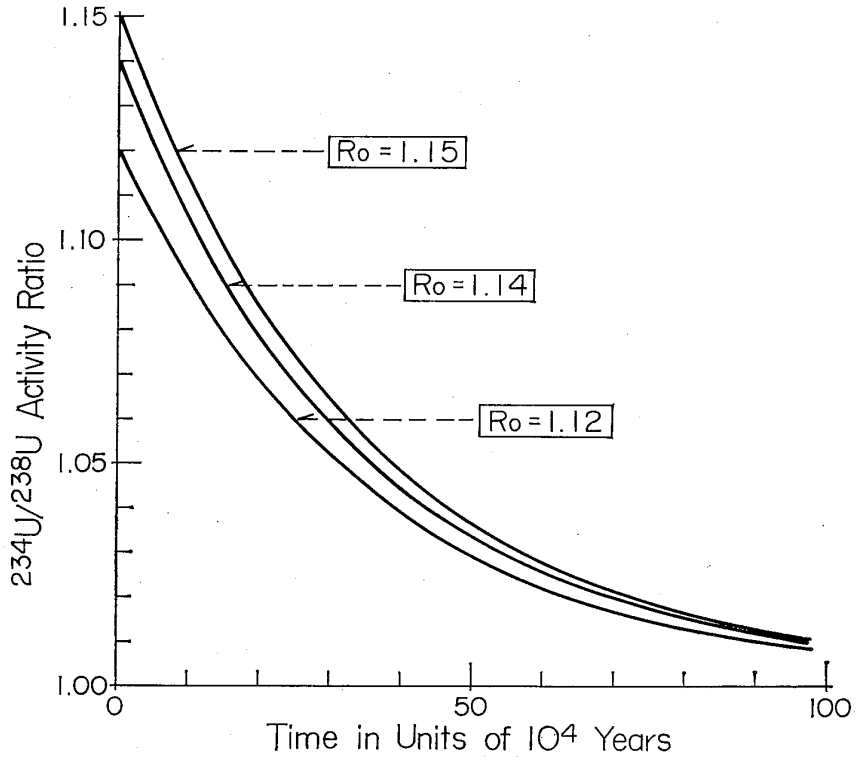

Fig. 3. Decay curves of the excess ^{234}U toward the secular equilibrium with ^{238}U for the initial $^{234}U/^{238}U$ activity ratios of 1.15 (Thurber, 1962), 1.14 (Ku *et al.*, 1977) and 1.12 (Nikolayev *et al.*, 1979).

years B. P. (Bloom *et al.*, 1974; and others). The then reefy limestone have been reported on Kikai-jima, which is located approximately 120 km NE of Tokuno-shima and is the outermost island in the central segment of the Ryukyu island arc, by Konishi *et al.* (1970, 1974). They argued from the present elevation of such a limestone unit and the other units formed during the subsequent interstadial phases that Kikai-jima has been rising at a rate of 1-2 m per 1,000 years since the last 120,000 years due to the neotectonic interplay as plate convergence of the active island arc and trench system. If Tokuno-shima has not been moved vertically during the past 120,000 years, the island should be surrounded by the coraliferous limestone unit which reaches the maximum elevation of about 6 m and is dated as being 120,000 years. As concluded above, even the uppermost part of the Pleistocene series is thought to be Middle Pleistocene in age and no Late Pleistocene reefy limestone is found on Tokuno-shima. Consequently, no occurrence of the reefy limestone formed in the last interglacial stage may be suggestive that Tokuno-shima locating more backward than Kikai-jima in the Ryukyu island arc, appears to have been subsided during the past 120,000 years. In addition, the absence of the Holocene Raised Coral Reef Limestone (Hanzawa, 1935) on Tokuno-shima may support that the vicinity of Tokuno-shima has been locally subsided since the last 120,000 years.

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徳之島“亀津層”（琉球石灰岩）の放射年代：南西諸島徳之島に分布する更新統最上部とされている亀津層（中川，1967）から採集したサンゴ（*Acropora* sp.）化石について，^{230}Th 年代を求めた。その結果，$^{230}Th/^{234}U$ 放射年代測定法の適用限界か，あるいはそれを越える値（30万年以上）が得られた。また $^{234}U/^{238}U$ 放射能比に基づく ^{234}U 年代は 387,000～709,000 年と推定された。すなわち，亀津層の年代値は30万年以上，そしてその上限は約70万年と結論される。結局，徳之島には喜界島などで見られる最終間氷期およびその後の数回の亜間氷期に形成された礁性堆積物（Konishi *et al.*, 1974）や完新世の“隆起サンゴ礁石灰岩”（Hanzawa, 1935, の Raised Coral Reef Limestone）が現在陸上に露出していなく，このことは，少なくとも過去12万年間は，徳之島付近が沈降傾向にあることを示唆している。　大村明雄
