Status and application of α-spectorometric 230Th/234U dating of fossil corals in Ryukyus, Japan and Philippinees

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Status and application of α-spectrometric ^{230}Th/^{234}U dating of fossil corals in Ryukyus, Japan and the Philippines

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Abstract : High-precision α-spectrometric ^{230}Th/^{234}U dating was achieved by recent improvements of measurement system and chemical procedures and enabled critical evaluation of age reliability. We review the status and application of α-spectrometric ^{230}Th/^{234}U dating of Pleistocene and Holocene corals to reconstruct past sea level changes and tectonic movements in Ryukyus, southwestern Japan and the Philippines in the western rim of circum-Pacific island arcs. The highest terrace in Kikai Island was formed during MIS 5c not MIS 5e that previously reported. Coral reef sediments deposited not only during MIS 5e but also during glacial periods, *e.g*. MIS 6 and 2, have been found in the Ryukyus. Coral reef sediments formed during MIS 2 were found at *ca*. 120 m below present sea level off Irabu Island located at 25°N. In addition, it was clear that three terraces developed during MIS 5e, 5c and 5a at Pamilacan Island on the Philippines.

1. Introduction

The ^{230}Th/^{234}U method of dating has been developed in the 1950s by using α-spectrometry (*e.g.* Barnes *et al.,* 1956) and has been advanced by the introduction of the mass spectrometric techniques, such as Thermal Ionization Mass Spectrometry (TIMS ; *e.g.* Edwards *et al.,* 1987) and Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS ; *e.g.* Stirling *et al.,* 2001). These mass spectrometric methods are powerful alternatives to the classical method of counting alpha decay to measure U-series radionuclides. The TIMS and MC-ICPMS ^{230}Th/^{234}U methods have succeeded to reduce both statistical errors of ages and sizes of samples for dating. These advantages have contributed to establish detailed sea level change during the last glacial cycle from fossil reefs (*e.g.* Chappell *et al.,* 1996 ; Gallup *et al.,* 1994) and to date terrestrial climate records in speleothems containing small amounts of uranium (*e.g.* Winograd *et al.,* 1992 ; Wang *et al.,* 2001). TIMS dating, however, has scarcely been used in Japan because of legal restrictions. Therefore, Omura *et al.* (1995a) developed high-resolution α-spectrometric ^{230}Th/^{234}U dating techniques that maximizes the potential of the traditional counting method. Here we present the status of the α-spectrometric ^{230}Th/^{234}U dating of fossil corals and its application for studies of coral reefs and reef terraces.

Fossil coral reefs are important as paleo-sea level indicators to unravel histories of sea level changes and tectonic movements during the late Quaternary particularly in the western Pacific Rim. Rates and patterns of tectonic uplifts have been deduced from elevation of reef terraces of Marine Isotope Stage (MIS) 5e in the Ryukyu and Daito Islands, southwestern Japan (*e.g.* Ota and Omura, 1992) and Huon Peninsula, Papua New Guinea (*e.g.* Chappell and Shackleton, 1986). However, the age of the MIS 5e terraces had been confirmed by the $^{230}Th/^{234}U$ dating only in five islands, Kikai, Hateruma, Yonaguni, and Kita- and Minami-Daito Islands until the mid 1990s in Japan (Fig. 1 ; Ota and Omura, 1992 and references therein), because it is difficult to collect unaltered fossil corals for dating. Further, the Philippines had been left as a blank area of age data of coral reef terraces until the 1990s in the western rim of circum-Pacific island arcs. Recently, there have been a number of studies on α-spectrometric $^{230}Th/^{234}U$ ages of reef terraces from Ryukyu Islands, Taiwan, and the Philippines (*e.g.* Yamada *et al.,* 2003 ; Yamaguchi *et al.,* 2004 ; Omura *et al.,* 2004). In addition, former $^{230}Th/^{234}U$ ages from Kikai Island were re-dated using high-resolution α-spectrometric $^{230}Th/^{234}U$ dating. We thus summarize recent studies in Kikai Island of central Ryukyus and the Philippines, and further mention important findings from a submerged reef western off Irabu Island, southern Ryukyus of Japan (Fig. 1).

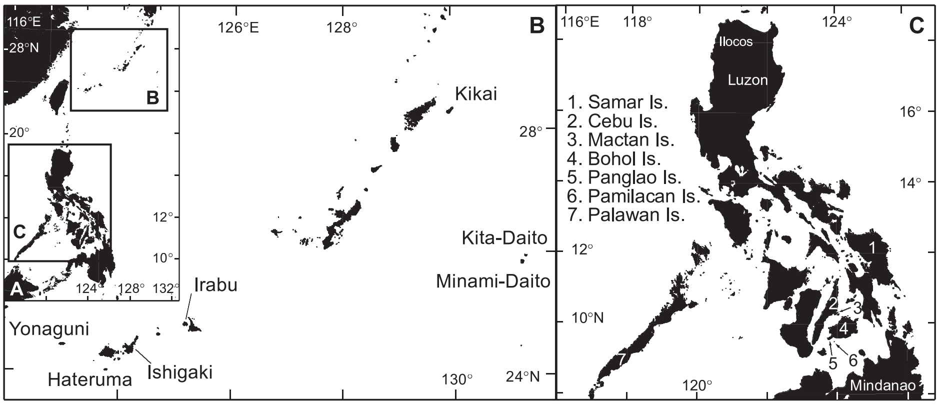

Figure 1. (A) Map showing the location of Ryukyu Islands, Taiwan and the Philippine Islands. (B, C) Enlarged map of Ryukyu Islands and the Philippine Islands, respectively.

2. Accuracy and precision of the α-spectrometric $^{230}Th/^{234}U$ method

The α-spectrometric method counts the numbers of alpha particles emitted from U and Th isotopes in contrast with the mass spectrometry, which measures the abundance of U-series radionuclides remained. Because the half-lives of ^{238}U, ^{234}U and ^{230}Th are long, it is essential for reducing age errors that the radioactivity of these nuclides is measured by α-spectrometry for prolonged periods. The high-resolution α-spectrometric $^{230}Th/^{234}U$ dating has been achieved by using both enlarged sample weights (up to 10 g) and prolonged counting time by the α-spectrometry (Omura *et al.,* 1995a). Such measurements have been reached by improvements in energy resolution and counting efficiency of the detectors. Age errors are less than 3% in 2σ range for MIS 5e corals using the recent α-counting system (*e.g.* Omura *et al.,* 1995a ; Inagaki and Omura, 2006). Solitary corals can also be dated with the same precision by using at least 2 g of samples.

Further, it has been possible to apply high-resolution α-spectrometric $^{230}Th/^{234}U$ method to Holocene corals with age errors less than 4% (Sasaki *et al.,* 1998 ; Ota *et al.,* 2000). The accuracy of α-spectrometric $^{230}Th/^{234}U$ ages of Holocene corals was confirmed by comparing with calibrated Accelerator Mass Spectrometric (AMS) ^{14}C ages of the same samples (Sasaki *et al.,* 1994). Fossil corals dredged from the continental shelf off southern Queensland, Australia, were dated to be from 11.25 to 6.36 ka by the $^{230}Th/^{234}U$ method (Sasaki *et al.,* 1994). These ages are consistent with cal. AMS^{14}C ages calibrated by using a well-established reservoir age of 450±35 yr (Gillespie and Polach, 1979). Thus, this method can be used as an effective alternative to ^{14}C because it does not require any correction for the reservoir effects and calibration for calendar ages.

To obtain reliable $^{230}Th/^{234}U$ ages, it is essential that coral samples satisfy the prerequisite conditions ; samples should initially be free of non-radiogenic ^{230}Th and have been preserved as a closed system for U and Th isotopes. After calculation of $^{230}Th/^{234}U$ ages, their reliability was evaluated using the five-step screening method with the following criteria (Omura *et al.,* 1995a, 2004 ; Sasaki *et al.,* 2006) : (1) samples have no evidence of re-crystallization of coral skeleton and/or cementation with secondary calcite or aragonite, (2) skeletal textures have not undergone changes through their diagenetic history, (3) the sample should not contain ^{232}Th, indicating free of initial and secondary contaminated ^{230}Th, (4) the uranium concentration should be the same as those in the present-day counterparts, ranging from 2 to 4 ppm, except for *Acropora* corals, which incorporate 3-4.5 ppm of U in their skeletons, (5) the initial $^{234}U/^{238}U$ activity ratio should be within 1.13-1.16 in median values, or should be consistent with that of the modern corals, 1.1466±0.0014 in 2σ ranges. Figure 2 shows a direct comparison of the high-resolution α-spectrometric $^{230}Th/^{234}U$ ages with the TIMS ages of the same coral specimen. Only ages satisfying these criteria (solid circles) are consistent with each other (Omura *et al.,* 1995b). This supports the accuracy of this method even with larger errors than those of the TIMS and the validity of the five-step screening method.

3. Application to coral reef studies in Kikai Island

Kikai Island is characterized by the highest uplift rate, ~1.8 m/ky, in the Ryukyu Islands (*e.g.* Ota and Omura, 1992). The main part of the island has three major Pleistocene terraces. The island is encircled with Holocene raised coral reef, which forms four steps of marine terraces (Sugihara *et al.,* 2003, and references therein). The elevation of the highest point in Kikai Island is 214 m on the topographic map (Fig. 3).

Many U-series dates of coral limestone in Kikai Island have been reported since the

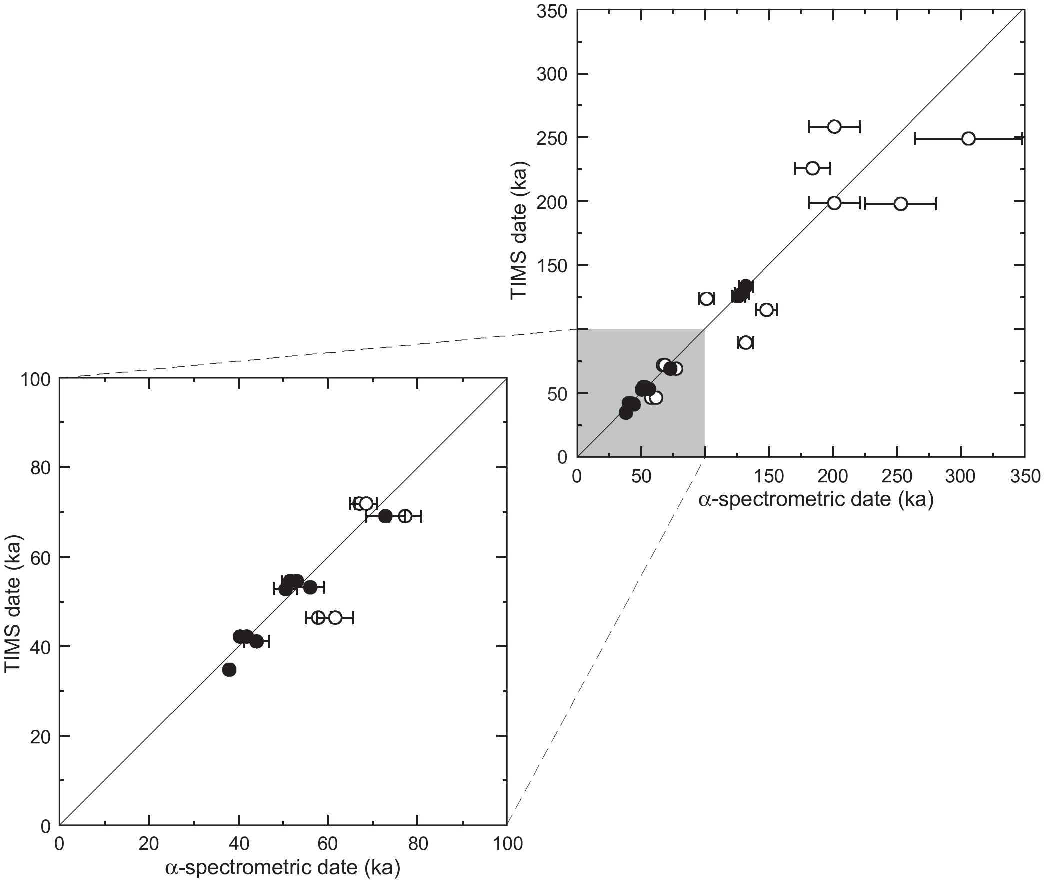

Figure 2. Comparison between α-spectrometric $^{230}Th/^{234}U$ and TIMS $^{230}Th/^{234}U$ dates of the same samples collected from Huon Peninsula, Papua New Guinea (Omura *et al.,* 1995b).
Solid and open circles represent reliable and less reliable dates based on the five step screening method.

1970s (*e.g.* Konishi *et al.,* 1974). Previous dates, however, were not so precise that reliabilities of ages could not be strictly evaluated based on the five-step screening criteria. Here, we introduce chronologic studies in Kikai Island within the past decade since the reliability and accuracy were improved.

3.1. Dating of solitary corals

Recent studies for the Ryukyu Group composed of Pleistocene coral reef sediments focus to distinguish each lithofacies, *e.g.* coral, rhodolith, detrital, and *Cycloclypeus-Operculina* limestone, as coeval components of coral reef complex and to regard one reef complex as one stratigraphic unit based on the modern biofacies and lithofacies around Ryukyu Islands (*e.g.* Iryu *et al.,* 1992,1998 ; Tsuji, 1993). This approach permits to interpret sedimentary succession associated with sea level change. However, it was difficult to

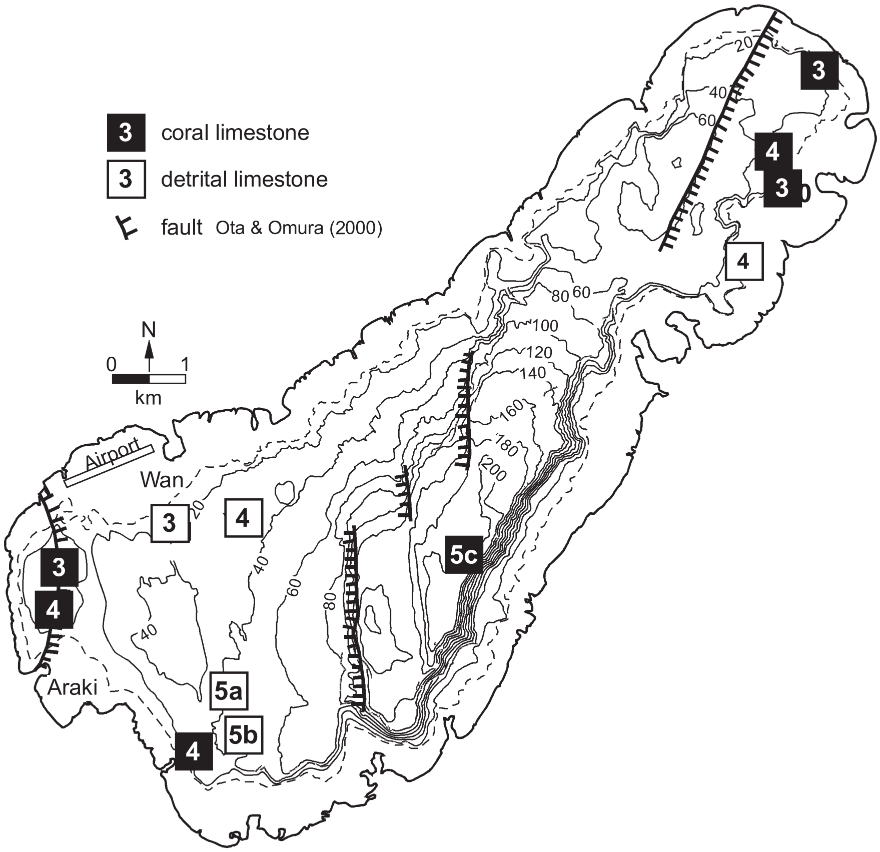

Figure 3. Topographic map of Kikai Island and distribution of dated Late Pleistocene coral reef sediments.
Number in boxes means marine isotope stage. Data were derived from Omura *et al*. (2000), Sasaki *et al*. (2004), Inagaki *et al*. (2005) and Inagaki and Omura (2006).

distinguish units containing detrital limestone because the depositional depth of this facies has wide range.

Since improvements of measurement system enable to date using small amount of sample, solitary corals can be dated accurately. The age obtained from autochthonous solitary corals indicate the depositional age of the host sediment. Thus, we can certainly distinguish units of reef complex based on high-resolution ages separately from unclear depositional depth.

Detrital limestone is extensively distributed on southwestern part of the lowest Pleistocene terrace. Omura (1983) reported that the detrital limestone was fore-reef sediments deposited during MIS 5a based on the U-series dating of solitary corals collected from the limestone. Inagaki *et al*. (2005) provided new U-series ages from solitary corals collected from detrital limestone at several sites on the terrace and concluded that the detrital limestone was deposited from 93 to 56 ka, corresponding to MIS 5b to 3 (Fig. 3). The detrital limestone was the prograded insular shelf sediments from MIS 5b to 3.

3.2. The highest Pleistocene terrace

Previous work concluded that the highest terrace and the coral limestone distributed on the cliff between the highest and middle terraces were formed during MIS 5e and 5c, respectively (*e.g.* Omura, 1988). However, these results were based only on dating and were not supported by litho- and bio-facies data.

Recent studies combining dating and litho- and biofacies observation show new information of the terrace age. Coral limestone collected from bindstone at *ca*. 195 m in elevation was dated as 104-97 ka, corresponding to MIS 5c (Fig. 3). This bindstone consists of an assemblage of *Porites* sp., *Favia speciosa, Cyphastrea* sp., *Goniastrea* sp., *Acropora palifera* (encrusting) and other hermatypic corals, indicating depositional depths of 5 – 15 m (Sugihara *et al.,* 1999 ; Sagawa *et al.,* 2001). The age correlated with MIS 5e was obtained from only one *Porites* sample, which is difficult to use as a depth indicator. These facts suggest that the highest terrace in Kikai Island was formed during MIS 5c not 5e (Inagaki and Omura, 2006). We presume that the relative sea level position during MIS 5e was situated at a higher position not just above the top of the island based on the litho- and biofacies. The uplift rate of Kikai Island since MIS 5e was re-calculated to be 2.1-2.3 m/ky using the new terrace age of MIS 5c (Inagaki and Omura, 2006). A coral limestone dated as MIS 6 was also found on the highest Pleistocene terrace. Exposure of MIS 5e or 6 corals is possibly due to erosional effect during MIS 5c transgression (Inagaki and Omura, 2006).

3.3. The lowest Pleistocene terrace

Chronological studies of coral limestone on the lowest Pleistocene terrace have indicated that the coral limestone was deposited at 65-50 ka, corresponding to MIS 3 (Omura, 1988). Konishi (1967) called the youngest Pleistocene coral limestone in Kikai Island as Araki Limestone, which was dated about 45-35 ka (Konishi, 1967 ; Konishi *et al.,* 1974 ; Omura, 1988).

During the same period, nine reef terraces were developed in Huon Peninsula (Omura *et al.,* 1995b ; Chappell *et al.,* 1996). To clarify the differences between Kikai and Huon reefs in the MIS 3, detailed stratigraphy was done using high-resolution α-spectrometric ^{230}Th/^{234}U dating. Newly obtained ages demonstrated offlapping sequence of five reef complexes, ages of which are ~66, ~64, ~62, ~55, and ~52 (Omura *et al.,* 2000 ; Sasaki *et al.,* 2004). Coral limestone deposited at ~46 and ~41 ka also distributed on the lowest Pleistocene terrace (Inagaki *et al.,* 2005). Four of these ages, ~62, ~52, ~46 and ~41 ka, can be correlated with the Huon terraces (Fig. 4). It was suggested that the reefs in Kikai were developed in response to the millennial scale climatic change (Sasaki *et al.,* 2004). No Pleistocene carbonate sediments, younger than about 41 ka, are exposed on the surface of Kikai Island (Inagaki *et al.,* 2005).

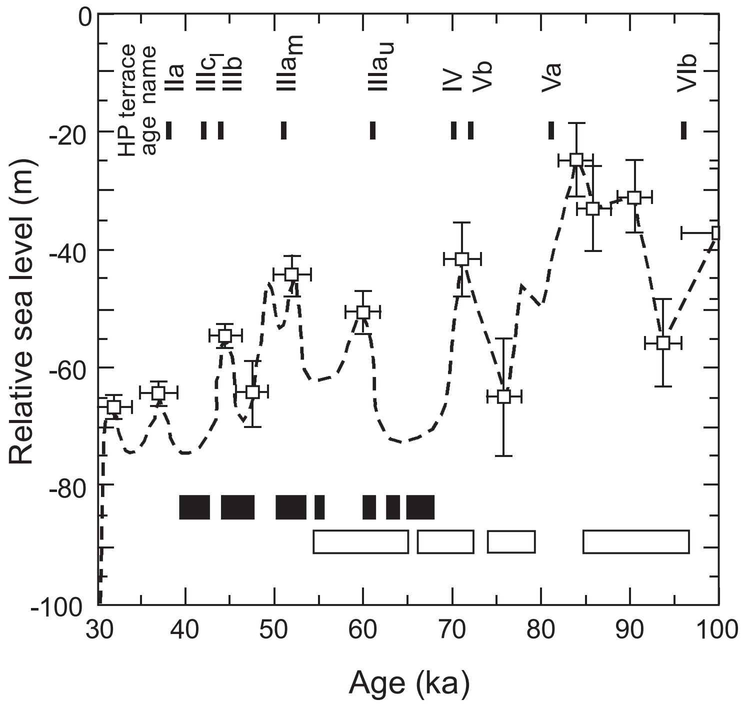

Figure 4. Relative sea level curve from uplifted coral reed terraces in Huon Peninsula, Papua New Guinea (HP) (Lambeck *et al.,* 2002) and ages obtained from the lowest Pleistocene terrace in Kikai Island.
Number and age of terraces are taken from Chappell *et al*. (1996). White and black boxes mean the age of detrital limestone and coral limestone, respectively (Omura *et al.,* 2002 ; Sasaki *et al.,* 2004 ; Inagaki *et al.,* 2005).

3.4. Holocene reef terraces

Recent studies of Holocene reefs in Kikai Island refined the timing and magnitude of uplift events and the growth pattern of the raised reef. Sugihara *et al*. (2003) found that a coral species, *Pocillopora verrucosa*, has a peak abundance at a depth of 1.5 m and is a reliable sea level indicator in the Ryukyu area. They reconstructed relative paleo-mean sea levels at 10.8-11.1, 8.5-8.9, 5.0-5.3, 4.0-4.3 and 1.9-2.5 m based on the distribution of *P. verrucosa*. These data combined with cal. ^{14}C and α-spectrometric $^{230}Th/^{234}U$ ages show that the four terraces were formed in response to repeated uplift events at 6.3, 4.1, 3.1 and 1.4 ka. Noteworthy from these results is that precisely defined magnitudes and timings based on the calendar ages of uplift events also support the time predictable model, *i.e*., idea of regularity in large earthquake recurrence (Shimazaki and Nakata, 1980). The well-developed raised fringing reef along the northeastern coast was investigated by drilling five cores through the Holocene section and by dating corals with α-spectrometric $^{230}Th/^{234}U$ method (Ota *et al.,* 2000). The Holocene reef, which is underlain by unconsolidated Plio-Pleistocene mudstone, is composed of two lithologic units with a total thickness of 25.8 m and a basal age of 9.56±0.15 ka. The upper limestone "reef" unit, which consists of a typically-zoned coral reef community, overlies a lower mixed-carbonate "bank" unit, a mixture of terrestrial grains derived from the basal mudstone and limestone and Holocene bioclastics. This growth pattern can be classified as a catch-up reef and is important to understand the development of Holocene reef terraces.

4. Coral reef terraces in the Philippine Islands

Coral reef terraces and emerged tidal notches are well developed along certain coastal stretches of the Philippines. However, the implications of uplifted marine terraces on the regional tectonic movements have not been fully understood until recently due to the lack of age control. Since a collaborative work between Japan and the Philippines on the chronology of Late Pleistocene and Holocene coral reefs started in 1998, MIS 5e coral reef terraces have been found in Palawan (western Philippines), Cebu, Bohol and Panglao Islands (central Philippines) at 2 to >14 m, except for the case of up to 27 m in Pamilacan Island, neighboring Panglao and Bohol Islands (Fig. 1, Table 1 ; Omura *et al.,* 2004 ; Ringor *et*

Table 1. Elevation of the MIS 5e sea levels in the western rim of the circum-Pacific island arcs and in tectonically stable areas in the Pacific.

Area	Locality	Elevation (m)	References
Western rim of the circum-Pacific island arcs			
Ryukyu Islands	Kikai Is.	220—270	Inagaki and Omura (2006)
	Ishigaki Is.	15	Yamada *et al.* (2003)
	Yonaguni Is.	30	Ota and Omura (1992)
	Hateruma Is.	41	Ota and Omura (1992)
Daito Islands	Kita-Daito Is.	12	Ota and Omura (1992)
	Minami-Daito Is.	10	Ota and Omura (1992)
Western Philippines	Palawan Is.	6.8	Omura *et al.* (2004)
Central Philippines	Mactan Is.	>5	Omura *et al.* (2004)
	Bohol Is.	3—8	Omura *et al.* (2004)
	Panglao Is.	2—>13	Omura *et al.* (2004)
	Pamilacan Is.	20—27	Ringor *et al.* (2004)
Huon Peninsula, Papua New Guinea	Tewai	440	Chappell and Shackleton (1986)
	Kanzarua	330	Chappell and Shackleton (1986)
Tectonically stable areas in the Pacific			
Hawaii	Oahu Is.	7	Szabo *et al.* (1994)
Western Australia	Cape Range	3-6	Stirling *et al.* (1998)

al., 2004). Elevations of these MIS 5e terraces are lower than those in other circum-Pacific island arcs, such as the Ryukyus and Huon Peninsula (*e.g.* Ota and Omura, 1992 ; Chappell and Shackleton, 1986), and are rather similar to those in the stable area, such as Hawaii and western Australia (*e.g.* Stiring *et al.,* 1998 ; Szabo *et al.,* 1994) (see Table 1). These vertical stabilities in western and central Philippenes can be observed in Holocene reef terraces and emerged tidal notches. Maeda *et al*. (2004) investigated these sea level indicators in Ilocos, Palawan and Samar coupled with ^{14}C and $^{230}Th/^{234}U$ dating to estimate local and regional tectonic uplifts (Fig. 1). Paleo-mean sea levels in Holocene are less than 2.9 m in Palawan and Samar in contrast with those of up to 6.7 m in Ilocos. The oldest age is ~8.2 ka in the $^{230}Th/^{234}U$ method in Ilocos. Such an old age of Holocene corals above the present sea level can only be found in highly uplifted area, such as in Kikai (8.1 ka ; Sumi, 1999MS) and Taiwan (8.6 ka ; Yamaguchi *et al.,* 2004). In addition to these regional trends, differential local uplift and tilting have been revealed to be influenced by localized tectonic activities (Maeda *et al.,* 2004 ; Omura *et al.,* 2004 ; Ringor *et al.,* 2004).

Further, Ringor *et al*. (2004) demonstrated three successive coral reef terraces of MIS 5e, 5c and 5a developed at 20-27 m, 9-13 m, and 3-6 m, respectively, in Pamilacan Island, and succeeded to estimate paleo-sea levels during MIS 5c and 5a to be 9-11 m and 6-9 m below present, respectively, assuming constant uplift rate of 0.18-0.21 m/kyr since MIS 5e. These paleo-sea levels are consistent with uplift-corrected reef data from Haiti, Barbados and Huon Peninsula (Chappell and Shackleton, 1986 ; Dodge *et al.,* 1983 ; Gallup *et al.,* 1994) as well as with submerged reefs from the tectonically stable Florida margin (Ludwig *et al.,* 1996).

5. Submerged reef off Irabu Island

Fossil reefs of the Last Glacial Maximum (LGM) period were reported only from the tropical region, such as Barbados and Tahiti (*e.g.* Fairbanks, 1989 ; Camoin *et al.,* 2001). Sasaki *et al*. (2006) reported the LGM coral reef from western off Irabu Island in the subtropical region of western Pacific (Fig. 1). High-resolution seismic reflection profiles delineated the distribution of mounded-shaped reflection beneath the insular shelf off Irabu Island, the Southern Ryukyus of Japan (Obata and Tsuji, 1992). A sediment core through one of these structures was recovered from seafloor at -118.2 m by offshore drilling by the Technology Research Center, Japan National Oil Corporation (presently Technology and Research Center, Japan Oil, Gas and Metals National Corporation). Lithology and the coral fauna in the core indicate that the mounded structure was composed of coral-algal boundstone, suggesting a 5-m thick coral reef. High-resolution α-spectrometric $^{230}Th/^{234}U$ dating coupled with cal. AMS^{14}C ages of corals indicates reliable ages ranging from 30.47±0.98 to 22.18±0.63 ka. This proves that such a submerged reef was formed during the lowstand stage of MISs 3 to 2 at 25° N.

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