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ANOMALOUS HIGH ^{14}C ACTIVITY FOUND IN RECENT CORALS FROM THE PHILIPPINES*

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ABSTRACT. The carbonate skeletons of small living corals collected in Spring 1981 from Cebu Island, the Philippines, had ^{14}C activities up to 147% of recent standard. Similarly high values were found in the carbonate structure of three large coral heads, where the ^{14}C content of six penetrating cores was measured. In these corals the activity of the outer parts grown since 1960 reached values as high as 155% (corrected for $\delta^{13}C = -25‰$) while the inner part grown from 1860 to 1950 had values of 106 to 110%. The ^{14}C content of corals should be ca 116% due to the atomic bomb effect and 95% before 1955. The samples were taken from the shore, exposed to tidal waters, so that local contamination is improbable. Organic samples collected from the same region showed normal ^{14}C activity.

INTRODUCTION

During a research stay on the Philippines in Spring 1981 primarily devoted to metabolism in tropical shallow waters and the growth of coral reefs, we collected samples of living corals or nearly recent corals. Our intent was to determine the atomic bomb effect on small corals with diameters up to 10 or 15cm. Algae and banana leaves were measured for comparison. Cores from large coral heads, diameters 2 to 3m, were drilled to investigate the stable isotopes ^{18}O and ^{13}C (Pätzold, 1984) and the ^{14}C content of the carbonates in the past. The ^{14}C activity of cores from beach rock at the shoreline and from offshore reefs was also measured.

METHODOLOGY

The samples came from the shore of Mactan, a small island near Cebu City in the center of the Philippines (10° 17′ N, 124° 0′ E) (Fig 1 A, B). The region is rather well protected against the open ocean, that is, the Pacific in the east and the South China Sea and the Celebes Sea to the west, by several islands, leaving only small inlets to the Camotes Sea and Mindanao Sea. Therefore, the waters surrounding Mactan Island are not influenced by ocean currents or upwelling streams.

The small corals were collected in the shallow water region up to 300m offshore. A further coral originates from Negros Island ca 50km west of Cebu Island. The originally colored corals were treated for one day with sodium hypochlorite solution to destroy and remove organic matter until the remaining carbonate skeleton was purely white. The corals were air-dried and then sent to the Kiel laboratory where they were washed with distilled water and dried at 50°C, partly in vacuum.

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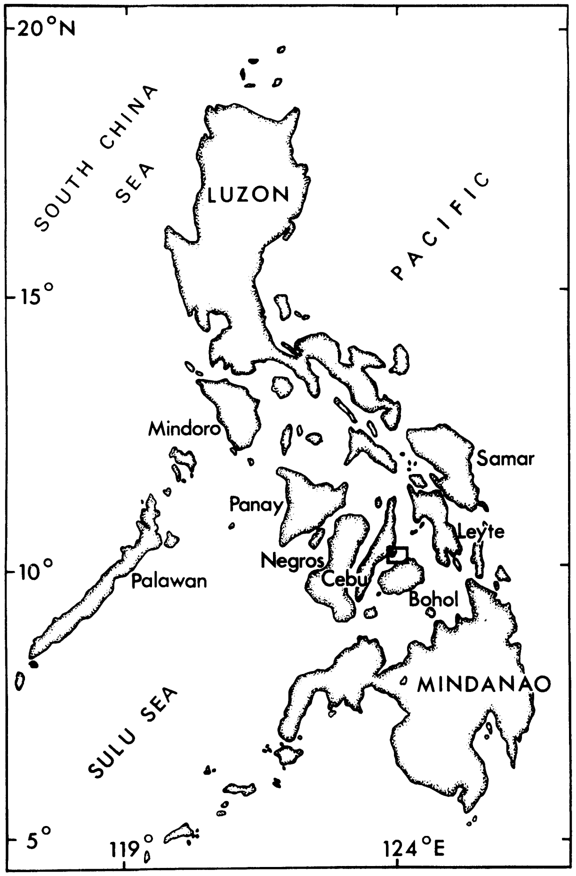

Fig 1A. Research area—The Philippines

DISCUSSION

The ^{14}C results are given in Table 1. All values are corrected to $\delta^{13}C$ = $-25‰$ according to

$$A_{corr} = A\left(\frac{1-\frac{25}{1000}}{1+\frac{\delta^{13}C}{1000}}\right)^2$$

Therefore, all results are directly comparable.

The activity of two organic samples, one terrestrial and one of marine green algae, were A = 128% and 111%, respectively, corresponding perfectly to values reported by Nydal, Lövseth and Skogseth (1980). These

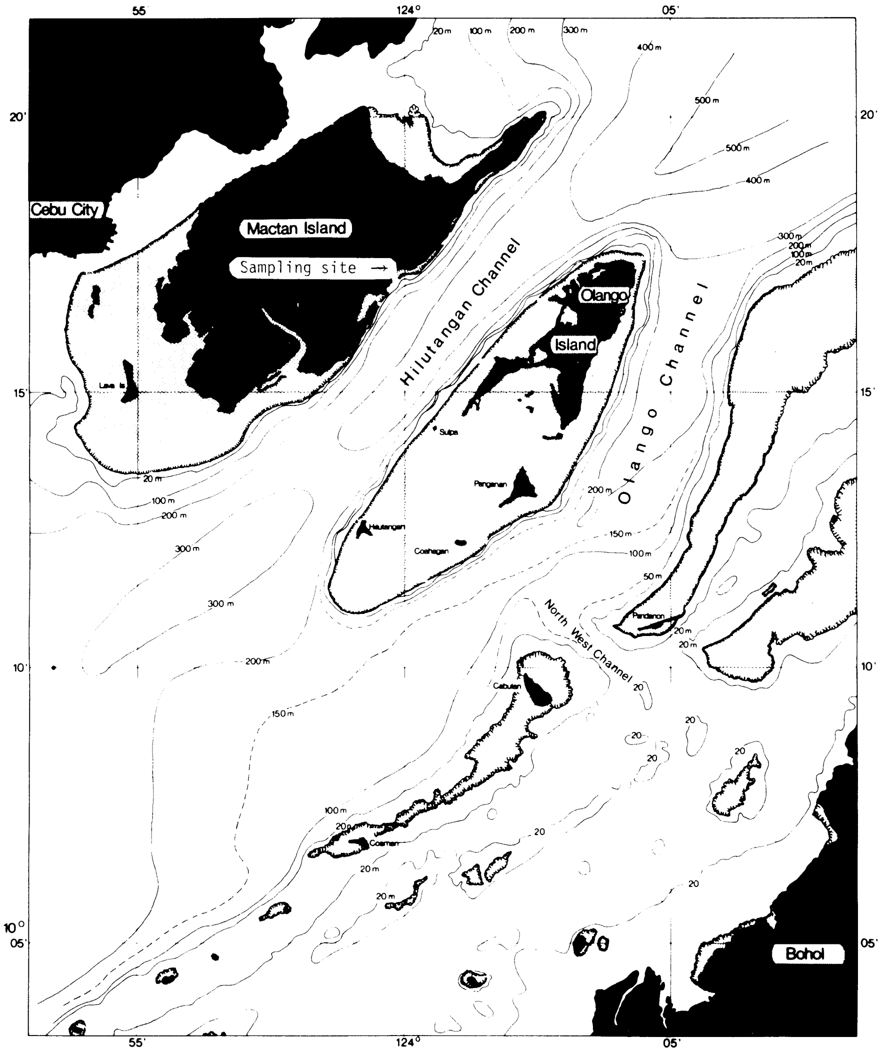

Fig 1B. Depth map of the region of Mactan Island (surveyed and drawn by Jan Rumohr, pers commun)

atmospheric values can be approximated by an exponential decay according to

$$\Delta^{14}C\% = 66 \cdot e^{-\lambda T}$$

$$T = \text{time since } 1.1.1967$$

$$\frac{1}{\lambda} = 16.7 \text{ yr}$$

TABLE 1
^{13}C and ^{14}C activity of small corals collected 1981 off Mactan Island, The Philippines

Sample no.	Description	$\delta^{13}C$	^{14}C activity (%) Measured value	^{14}C activity (%) Corrected to $\delta^{13}C = -25‰$
Leaves				
KI-1892	Banana leaf	−26.84	127.60 ± .55	128.07 ± .55
Algae				
KI-1891	Green algae	−16.73	112.70 ± .47	110.81 ± .46
Corals				
KI-1974	Tips of *Montipora fragilis* grown 1980, partly 1979	Mean value		137.27 ± .41
1974.011		− 2.66	142.76 ± .62	136.43 ± .59
1974.021	Dried 1 day in vacuo at 50°C	− 1.72	144.78 ± .59	138.10 ± .56
KI-2025	*Montipora ramosa* (whole stock, grown ca 1975–1980)	− 1.70	153.68 ± .48	146.59 ± .45
KI-2062	*Cycloseris sibogae* (Mushroom coral) small specimen large axis 28, small axis 25mm	− 1.85	119.94 ± .48	114.44 ± .46
KI-2063	*Galaxea clavus* (cut, grown ca 1977–1980)	− 2.47	144.72 ± .53	138.26 ± .51
KI-2064	*Coeloseris mayeri*	− 2.51	137.95 ± .52	131.80 ± .50
KI-2123	*Montipora ramosa* (whole stock) grown ca 1982, coll Feb 1983 off San Carlos City, Negros	− 4.49	117.50 ± .86	112.71 ± .82

For Spring 1981, this formula gives $\Delta^{14}C = 28\%$, the value found in the leaf. Likewise, $\Delta^{14}C = 11\%$ falls into the ocean range, and we see that for the late 1970s and more so for the following years, any value higher than $\Delta^{14}C = 16\%$ is improbable.

Instead of this, most corals had activities between A = 130 and 150%. Only a mushroom coral and the coral collected one year later off San Carlos, Negros Island, had normal values of 114 and 113%.

We then measured the ^{14}C content of coral head cores. The corals were drilled in three directions–vertical, parallel, and perpendicular to the coast (Grobe, Wefer & Willkomm, 1984). The cores, 3.5cm in diameter, were only slightly dried and sent to the Kiel Geologic Institute, where they were cut into slices (Fig 2). On the middle cut, the seasonal variations of density could be observed by light and X-ray photographs, and the annual variations were confirmed by a tight sequence of $\delta^{18}O$ measurements (Pätzold, 1984). Thus, we had well-dated material for the ^{14}C analysis, in one coral, dating back to 1860, and in two others, back to 1920.

The results, calculated for $\delta^{13}C = -25‰$, are given in Table 2 and plotted in Figure 3. The standard deviation, not shown in the diagram, lies between 0.4 and 0.6%. The continuous line repeated in all three figures is the medium value calculated from 4 cores of 2 coral heads and is used only as visual aid. For comparison, the dotted line is drawn representing the values of Druffel (1980), Druffel and Suess (1983), and Nozaki *et al* (1978) in the Caribbean and Bermuda.

Obviously, the material is contaminated, but how? Contamination in

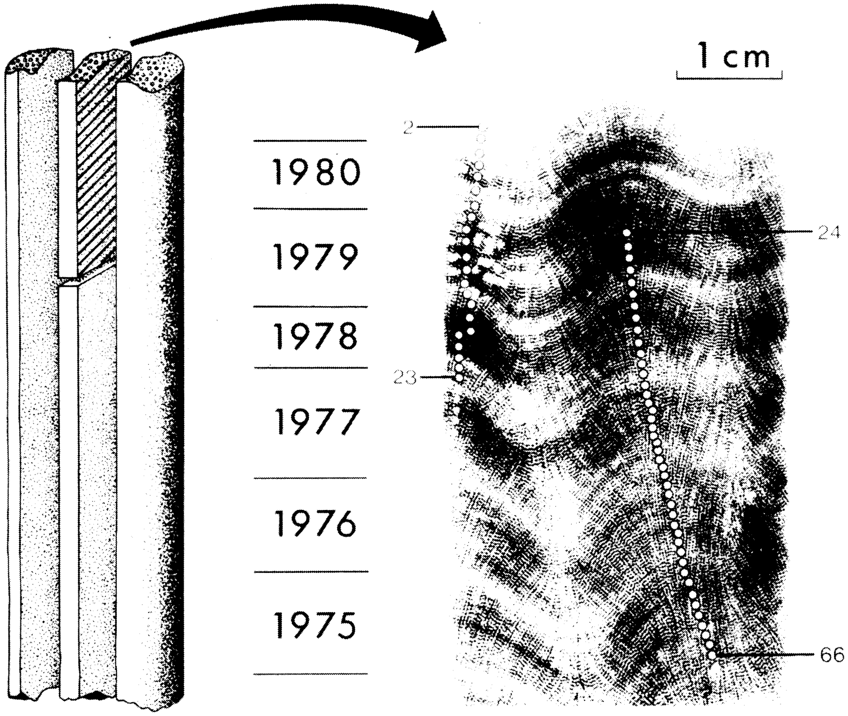

Fig 2. Cutting of coral cores and position of samples for stable isotope analysis (from Pätzold, 1984)

the laboratory is improbable for several reasons:

1) Samples from beach rock, that is, abiotic precipitated calcareous rock at the present or former shoreline, gave activities of 7 to 10%, corresponding to 20,000 BP (see Table 3). Likewise, deeper parts of cores from the Olango Flat, an old reef that dried up during the glacial period, gave ^{14}C ages between 16,000 and 25,000 BP (Grobe, Wefer & Willkomm, 1984). These samples were recovered and treated like the coral cores, but due to their total activity, the contamination cannot be higher than a few per cent.

2) The small corals were treated in a totally different way before measuring their activity, but the ^{14}C content is the same as in the large corals.

3) A further reason could be found in the excess activity, itself. If the additional ^{14}C content is caused by artificial ^{14}C, it is hard to explain why the excess just lies in the low range of 10 to 40% of standard recent activity. Moreover, the additional activity, compared to the values of the Caribbean, increases slowly between 1860 and 1960 from 12 to 16% and then goes up to 25 to 40%. An accidental contamination would not have been so regular.

TABLE 2

^{13}C and ^{14}C activity of cores from coral heads (*Porites*) of Mactan Island, The Philippines

Sample no.	Depth from coral surface (cm)	Growth year	$\delta^{13}C$‰	^{14}C content $\pm 1\sigma$ corrected to $\delta^{13}C = -25$‰
South Coral, Core IV to NNE				
KI-1951.01	0–5.5	1975/81	−1.31	138.5 ± 0.51
.03	14–18.5	1968/70	−1.64	144.6 ± 0.42
.05	20–29	1962/67	−2.25	124.3 ± 0.59
.09	44–48	1948/51	−1.68	111.5 ± 0.45
.10	57–61	1936/43	−1.13	111.0 ± 0.48
.14	99–103	1901/06	−0.52	107.8 ± 0.46
.22	199–203	ca. 1860	+0.37	106.9 ± 0.38
South coral, Core III to ESE				
KI-1950.01	0–6	1975/81	−3.12	133.9 ± 0.48
.012	6–12	1971/74	−2.31	133.9 ± 0.49
.02	15.5–21	1968/70	−3.1	119.1 ± 0.47
.03	23–29	1957/61	−2.02	112.9 ± 0.46
.05	41–48	1946/51	−1.60	109.4 ± 0.48
.07	52.5–58.5	1936/44	−1.59	109.1 ± 0.52
.10	69.5–76	1924/27	−0.63	111.4 ± 0.75
.14	120–127.5	1900/05	+1.19	105.6 ± 0.54
.20	202–209.5	1892/95	−0.44	111.2 ± 0.57
South Coral, Core II vertical				
KI-1949.01	4–9	?	−0.62	108.5 ± 0.75
.05	36–41.5	?	−0.48	104.8 ± 0.44
North coral, Core IX to NNE				
KI-2020.01	0–8	1970/80	−2.58	113.7 ± 0.46
.02	14–22.5	1958/65	−2.14	109.2 ± 0.57
.03	23.5–30	1950/56	−2.17	99.7 ± 0.54
.06	52–57.5	1929/33	−1.73	95.1 ± 0.44
.282	0–2	1980/81	−2.91	154.8 ± 0.68
.283	2–4.5	1978/79	−2.10	154.1 ± 0.51
Hadsan coral, Core XIV to E				
KI-1976.01	0–6.5	1977/81	−0.90	137.6 ± 0.49
.05	30–33	1958/60	−1.18	111.6 ± 0.46
.08	48–54	1943/48	−1.14	107.9 ± 0.45
.17	139–144.5	1966/69	−2.36	129.2 ± 0.61
Hadsan coral, Core XIII vertical				
KI-1975.01	11–17	1967/70	−0.77	129.1 ± 0.47
.05	44–50.5	1948/51	−0.74	116.4 ± 0.45
.06	64–70.5	1936/40	−0.60	113.8 ± 0.45
.09	88–95.5	1920/25	+0.90	105.8 ± 0.39

CONCLUSIONS

We must suppose a contamination in situ. Now, the main problem is the large distribution, locally as well as in time.

The distance between the north and south coral is only some 20m. But the small corals were collected in an area of some 100m around these coral heads. And the Hadsan coral with exactly the same activity as the south coral lies ca 1.6km southwest of the other corals. Thus, the contamination, whatever the reason, must have affected a rather large area. Also because of the tidal exchange of water, the excess activity cannot be confined to a small area. The most obvious explanation would be the atomic bomb effect. But published values (see eg, Linick, 1980) indicate that ^{14}C activity of sur-

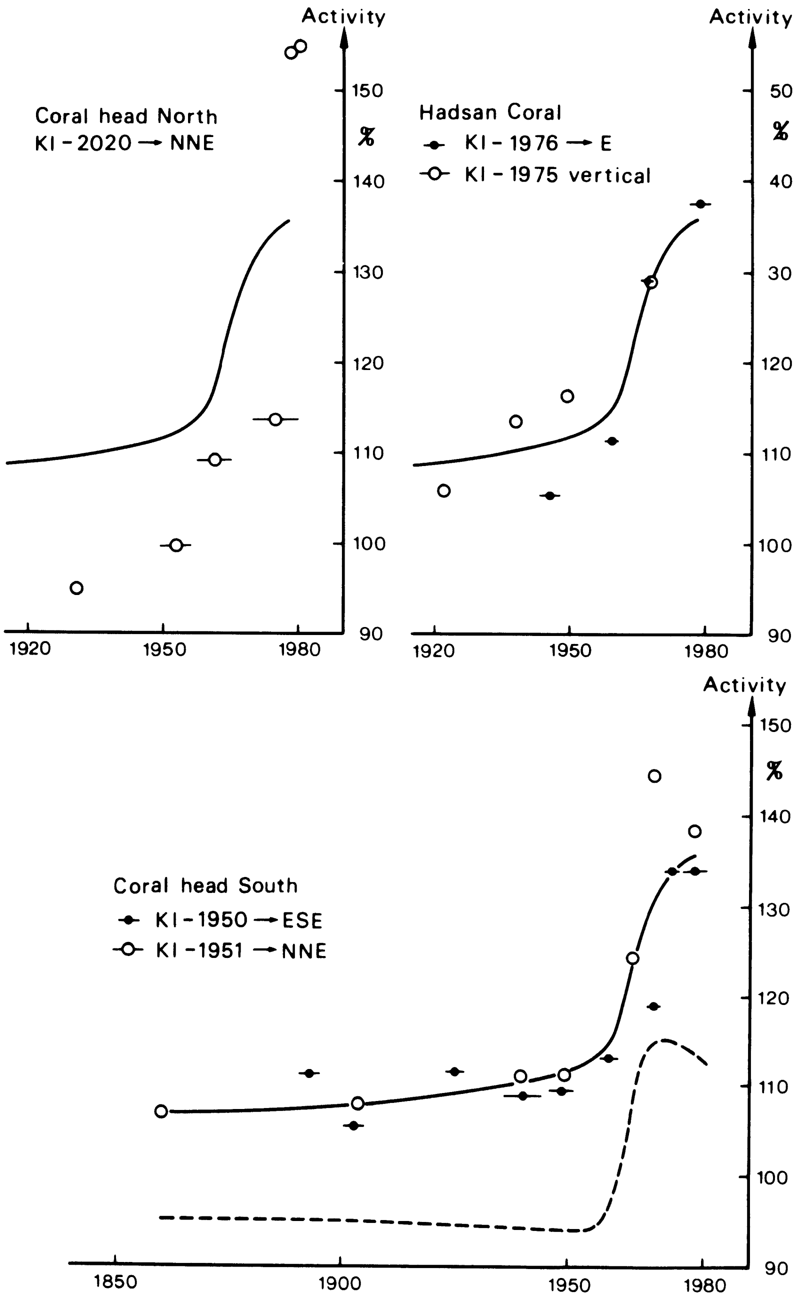

Fig 3. ^{14}C content of coral heads vs growth time. Activity is corrected to $\delta^{13}C = -25‰$, but not for age.

TABLE 3
^{14}C measurements on beach rock and coral reefs off Mactan Island, The Philippines

Sample no.	Drilling depth (cm)	$\delta^{13}C$	^{14}C activity ($\pm 1\sigma$) corrected to $\delta^{13}C = -25‰$	^{14}C age ($\pm 1\sigma$)
Beach rock				
KI-1897*	Surface	−4.26	6.88 ± 0.28	21,500 ± 320
KI-1965.02	Surface	+0.26	9.75 ± 0.33	18,700 ± 260
.15	125	−4.32	7.14 ± 0.34	21,200 ± 370
Slope of the reef				
KI-1966.04	30	−1.14	75.76 ± 0.43	2230 ± 43
KI-1967.01	Surface	+2.53	114.5 ± 0.57	———
.17	170	+2.27	91.7 ± 0.57	700 ± 47
KI-1968.02	Surface	+1.40	97.8 ± 0.51	180 ± 42
KI-1969.03	Surface	———	100.16 ± 0.44	———

* KI-1897: Cut at Mactan, ca 20m from shore at 4m asl
KI-1965: Core I, at the shore, vertically drilled into the beach rock, sea level
KI-1966: Core V, ca 300m offshore at 5m water depth, vertically drilled into the reef slope
KI-1967 to -1969: Cores VI, X, and XI; Core VI, ca 300m, Cores X and XI ca 100m offshore at 5m water depth, horizontally drilled into the reef slope

face waters in the Pacific never exceeded A = 120% or $\Delta^{14}C$ = 200‰. The normal activity of the algae also argues against this assumption.

The emission of ^{14}C nuclei from the decay of natural radioactive families (^{238}U, ^{235}U, and ^{232}Th) is currently being studied as an additional source for ^{14}C (see eg, Barker, Jull & Donahue, 1985, and references therein). In uranium minerals, the authors found a concentration of $2.2 \cdot 10^{-15}$ ^{14}C atoms/uranium atom. If the coral contains 10% of its weight as uranium, then the concentrations to expect are

$$\frac{1}{10} \cdot \frac{6 \cdot 10^{23}}{238} = 2.5 \cdot 10^{20} \frac{\text{U atoms}}{\text{g}}$$

and

$$2.5 \cdot 10^{20} \cdot 2.2 \cdot 10^{-15} = 5.5 \cdot 10^{5} \frac{^{14}\text{C atoms}}{\text{g}}$$

The decay rate of this uranium-born ^{14}C would be $1.3 \cdot 10^{-4}$ dpm/g coral or $1.2 \cdot 10^{-3}$ dpm/g C, which is <0.1‰ of standard recent activity.

The distribution in time is also difficult to understand. The inner parts of two coral heads grown before 1956 have an activity of 107 to 113%, thus exceeding even the atmospheric ^{14}C content. Therefore, the additional ^{14}C probably diffused into the already existing coral skeleton. Though the value of 120 to 150% of standard activity is extremely high for natural samples, the excess measured in normal radioactivity units is not exciting. The south coral, eg, may have a mass of $16 \cdot 10^{6}$g carbonate or $2 \cdot 10^{6}$g C. With a mean excess activity of ca 18% or 1.1pCi/g, the additional activity of the whole coral head is $\lesseqgtr 2$ μCi, a value small to activities used in biological tests with artificial radionuclides.

If the ^{14}C entered into the sea water during experiments, the total activity scarcely will have exceeded the order of some 100μCi and it must have infiltrated into the coral within one or a few tides. It is astonishing, then, that the ^{14}C penetrated nearly homogeneously the whole coral of ca 10m^3 within this comparatively short time, and the question remains how nearly the same activity came to a coral 1.6km away.

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REFERENCES

Barker, D, Jull, A J T and Donahue, D J, in press, Excess C-14 abundances in uranium ores: possible evidence for emission from uranium-series isotopes: Geophys Research Letters.

Druffel, E M, 1980, Radiocarbon in annual coral rings of Belize and Florida, *in* Stuiver, M and Kra, R S, eds, Internatl ^{14}C conf, 10th, Proc: Radiocarbon, v 22, no. 2, p 363–371.

Druffel, E M and Suess, H E, 1983, On the radiocarbon record in banded corals: Exchange parameters and net transport of $^{14}CO_2$ between atmosphere and surface ocean: Jour Geophys Research, v 88, no. C2, p 1271–1280.

Grobe, H, Wefer, G and Willkomm, H, in press, Internal structure of the double reef of North Bohol and the Olango Reef Flat (Philippines): Philippine Sci, v 21.

Linick, T W, 1980, Bomb-produced carbon-14 in the surface water of the Pacific Ocean, *in* Stuiver, M and Kra, R S, eds, Internatl ^{14}C conf, 10th, Proc: Radiocarbon, v 22, no. 3, p 599–606.

Nozaki, Y, Rye, D M, Turekian, K K and Dodge, R E, 1978, A 200 year record of carbon-13 and carbon-14 variations in a bermuda coral: Geophys Research Letters, v 5, no. 10, p 825–828.

Nydal, R, Lövseth, K and Skogseth, F H, 1980, Transfer of bomb ^{14}C to the ocean surface, *in* Stuiver, M and Kra, R S, eds, Internatl ^{14}C conf, 10th, Proc: Radiocarbon, v 22, no. 3, p 626–635.

Pätzold, J, 1984, Growth rhythms recorded in stable isotopes and density bands in the reef coral *Porites lobata* (Cebu, Philippines): Coral Reefs, v 3, p 87–90.