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Anomalous levels of ^{90}Sr and $^{239,240}\text{Pu}$ in Florida corals: Evidence of coastal processes

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Abstract—Strontium-90, a radionuclide whose primary source is fallout from nuclear weapons testing, serves as a tritium-like tracer of ocean circulation. The historical record of ^{90}Sr activities in the annual bands of island corals have been shown by other investigators to reflect the ^{90}Sr concentration in surface waters at those site. Strontium-90 activities measured in annual bands in *Montastrea annularis* from the Florida Keys are 30–120% higher than those in corresponding peak activity years (1960–1965) of a Bermuda coral (*Diploria*). The Bermuda ^{90}Sr activity record reflects the fallout source only, whereas the additional ^{90}Sr activity in the Florida Keys is expected to reflect a coastal runoff source as well as the fallout. The coastal circulation patterns off the northern and western edge of the Florida Current further act to concentrate and prolong the exposure of the runoff ^{90}Sr to the corals. Six measured $^{239,240}\text{Pu}$ activities in the Florida coral are 30% of $^{239,240}\text{Pu}$ activities in island coral records previously reported. Since Pu is expected to be scavenged by particles in coastal waters, this decrease in $^{239,240}\text{Pu}$ substantiates the importance of coastal influences in the Florida ^{90}Sr record.

Strontium-90 activities measured in subannual coral bands from 1973 to 1974 reflect seasonal changes in the ^{90}Sr concentrations in the surface layer of the coastal waters. This may reflect Loop Current intrusion events. The seasonal and long-term coral ^{90}Sr data presented in this paper suggests that coastal ^{90}Sr coral time series may be very useful for documenting coastal circulation patterns.

INTRODUCTION

THE TIME HISTORY of $^{14}\text{C}/^{12}\text{C}$ in atmospheric CO_2 using annual bands in tree rings was demonstrated by DEVRIES (1958) and many others. Similar records have been obtained from banded corals to track the history of changes in the $^{14}\text{C}/^{12}\text{C}$ ratio in the surface ocean (NOZAKI *et al.*, 1978; DRUFFEL and LINICK, 1978; DRUFFEL, 1981). Annual banding in corals is manifested by regular density changes in the accreted aragonitic skeleton. KNUTSON *et al.* (1972) was first to show that X-rays of slabs of coral cut along the vertical axis of growth of the scleractinian coral *Montastrea annularis* revealed consecutive dark and light bands. Sectioning the coral into yearly samples retrieves an historical record of the chemical and isotopic composition of the seawater surrounding the corals. The accreted skeleton of the coral has been shown to incorporate minor (*i.e.*: Sr, Mg, Na; WEBER, 1973, 1974; GOREAU, 1977; SMITH *et al.*, 1979; SWART, 1981) and trace elements (*i.e.*: U, Pu, Pb, Cd, Zn; LIVINGSTON and THOMPSON, 1971; BENNINGER and DODGE, 1986; SHEN and BOYLE, 1988; SWART and HUBBARD, 1982) with constant distribution coefficients (K_D) between seawater and skeletal concentrations (GOREAU, 1977; SWART, 1981; BENNINGER and DODGE, 1986).

Strontium-90 was introduced to the environment as a product of atmospheric nuclear weapons testing during the period 1952 to 1962. Unlike ^3H and ^{14}C , there is no natural source of ^{90}Sr . It is an excellent tracer of ocean circulation due to its conservative behavior in the sea and equally important, its incorporation into corals. There is no fractionation of $^{90}\text{Sr}/\text{Sr}$ in corals relative to its composition in seawater (TOGGWEILER, 1983; NOSHKIN *et al.*, 1975) and the Sr/Ca ratio in corals relative to seawater is affected to only a minor extent by sea surface temperature, coral growth rates and coral species (THOMPSON and LIVINGSTON, 1970; WEBER, 1973; GOREAU, 1977; HOUCK *et al.*, 1977; SMITH *et al.*, 1979;

SWART, 1981; SCHNEIDER and SMITH, 1982). For example, BENNINGER and DODGE (1986) reported the constancy of Sr concentration per gram of coral over a 30 year period in *Montastrea annularis* from 1951–1980, with a $K_{D(\text{Sr})} = 1.040 \pm 0.008$.

The feasibility to recover ^{90}Sr records from corals and the similarity of these records to the fallout ^{90}Sr atmospheric record as measured in New York City was documented by TOGGWEILER (1983) in corals from the Atlantic (Bermuda) and the Pacific (Oahu) (TOGGWEILER and TRUMBORE, 1985). Measuring ^{90}Sr activity levels in corals has the advantage of providing a complete long-term historical record of the ^{90}Sr activity present in the waters at one site. Previously, only limited time-series measurements were made for locations throughout the Atlantic and Pacific during seawater sampling programs, but these measurements could not represent the dynamic changes occurring constantly throughout the oceans. In addition, annual sampling of coral bands average out seasonal changes which would be reflected in ^{90}Sr levels measured in single seawater samples.

Sr removal from the atmosphere is much faster than removal of CO_2 , thus the response time of the oceans to fallout ^{90}Sr is on the order of one year *versus* 10 years for ^{14}C . Although bomb-generated ^3H has been used as a tracer for ocean circulation (*e.g.*, OSTLUND *et al.*, 1974; JENKINS, 1980; JENKINS and CLARKE, 1976; FINE *et al.*, 1981), future ^3H measurements will be limited by the sensitivity of the analytical instrumentation used for ^3H due to its short half-life of 12.4 years. Presently there is no practical method for retrieving the ^3H record from the organic matter in corals. Strontium-90's half-life of 28.5 years, combined with the input record in corals, provides a complementary tracer whose potential has not yet been fully realized.

In order to use ^{90}Sr in corals as an effective circulation tracer, the input function of ^{90}Sr to the coral site must be known. For instance, corals growing in coastal areas may be

influenced by fluvial transport of ^{90}Sr from the continents, in addition to the fallout input. Previous ^{90}Sr coral work (TOGGWEILER, 1983; TOGGWEILER and TRUMBORE, 1985) has focused on samples from central gyres, far from continents, where the input was dominated by direct deposition from the atmosphere. In contrast, this paper concerns the historical record from the Florida Keys, a near shore site. The results are compared with those from a Bermuda coral representative of a central gyre site. The unexpectedly high ^{90}Sr concentrations in the Florida coral as compared to the Bermuda coral suggest strong coastal influence in the Florida coral record. Extremely low $^{239,240}\text{Pu}$ concentrations in Florida coral relative to a Caribbean coral (BENNINGER and DODGE, 1986) further confirm our suspicion of coastal influence.

COLLECTION OF CORALS AND ^{90}Sr AND $^{239,240}\text{Pu}$ ANALYSIS

Cores from massive colonies of *Montastrea annularis* were collected in 1983 from "The Rocks" reef ($24^{\circ}57'\text{N}$, $80^{\circ}33'\text{W}$) off the Florida Keys from 4 meters water depth (see Fig. 1). This coral reef is 2 km

south of Plantation Key. The major water mass to the south of these corals is the Florida Current, which is part of the Gulf Stream system. These corals are influenced by the surface waters of the Florida Current and the Florida Bay to the north. "The Rocks" samples were obtained from one core (TR3) collected in February, 1983; it contained growth from A.D. 1939 to 1983.

An additional specimen of *Montastrea annularis* from New Ground ($24^{\circ}40'\text{N}$, $82^{\circ}25'\text{W}$) was collected in 4.0 meters in July, 1983 (see Fig. 1). This site was chosen to represent a second distant Florida Keys reef site that might be influenced by runoff from the land to provide a check of ^{90}Sr activities recorded in the first primary Florida samples. The New Ground coral was living in green, "murky" water, a sign of coastal influence instead of the dark, purple-blue waters of the Gulf Stream (E. SHINN, pers. commun.). Bands representing four years were selected from this coral, although the 1960 sample was lost.

The Bermuda ^{90}Sr results were obtained from *Diploria strigosa* cored from a 1 m diameter colony at North Rock located 15 km north of Bermuda at 10 m depth. This core was collected in August of 1983 and contained growth for the period A.D. 1882 to 1983. The growth rate of this coral was only 3–6 mm per year, much slower than *Montastrea annularis* at Florida (8–10 mm/yr). Eight annual bands were analyzed from the period 1951 (^{90}Sr blank) to 1982.

All coral cores were slabbbed along the axis of growth (1 cm thick), ultrasonically cleaned, X-rayed and mapped (GRIFFIN and DRUFFEL,

FIG. 1. Sites of coral collection: (1) North Rock, Bermuda (10–15 miles north of Bermuda) in 10 m of water on August 25, 1983, (2) The Rocks reef, Florida Keys (off Plantation Key) in 4 m of water in March, 1983, inset A, (3) New Ground reef, Florida Keys (35 miles west of Key West) in 4 m of water on July 28, 1983, inset B. Size of arrow does not reflect volume transport, merely general direct of currents.

Table 1: ^{90}Sr and $^{239,240}\text{Pu}$ Activities in *Montastrea annularis* from two Florida Key sites and in *Diploria strigosa* from North Rock, Bermuda.

Year	^{90}Sr Florida The Rocks reef dpm/100g coral	$^{239,240}\text{Pu}$ Florida The Rocks reef dpm/kg coral	^{90}Sr Florida New Ground reef dpm/100g coral	^{90}Sr Bermuda North Rock reef dpm/100g coral
1940	0.0 $\pm$ 2.6	0.007 $\pm$ 0.014		
1947	7.3 $\pm$ 5.5			
1951	4.1 $\pm$ 0.6			0 $\pm$ 2.3
1952	lost		0 $\pm$ 3.1	
1955	17.5 $\pm$ 0.3			
1957	22.9 $\pm$ 1.2	0.77 $\pm$ 0.18		
1958	33.7 $\pm$ 1.4			
1960	41.5 $\pm$ 2.1		lost	18.6 $\pm$ 3.0
1961	21.9 $\pm$ 1.1	0.85 $\pm$ 0.21		
1962	31.2 $\pm$ 0.7	0.85 $\pm$ 0.15		24.1 $\pm$ 4.3
1963	55.5 $\pm$ 0.9	1.86 $\pm$ 0.25		
1964	100.5 $\pm$ 1.1	3.68 $\pm$ 0.29	97.3 $\pm$ 2.0	55.0 $\pm$ 1.3
1965	74.8 $\pm$ 0.8			49.8 $\pm$ 1.4
1966	57.1 $\pm$ 1.1	0.47 $\pm$ 0.08		
1967	48.3 $\pm$ 1.5	0.87 $\pm$ 0.16		
1968	48.1 $\pm$ 1.7*			
1970	37.1 $\pm$ 1.9			28.1 $\pm$ 1.4
1973	20.9 $\pm$ 1.8**			
1974	17.9 $\pm$ 1.2**		23.2 $\pm$ 1.7	26.4 $\pm$ 0.9
1978	17.9 $\pm$ 0.3			
1982	12.7 $\pm$ 0.2			12.4 $\pm$ 2.1

* Average for two samples

** Average for four seasons

1985). Each annual sample was sectioned from the core by cutting (with a band saw) through the center of the dense band in *Montastrea annularis* from Florida; in this way, a given band for year x represented the growth interval from August of year $x - 1$ to July of year x . For the *Diploria strigosa* samples, the cut was made just below the dense band, representing growth from April to March. Seasonal samples were sectioned from two years (1973 and 1974) in "The Rocks" Florida coral by grinding three-month intervals using a Dremel tool. Annual coral bands were pulverized and dissolved in 8 N HNO_3 acid. The 1973, 1974, 1978, and 1982 Florida (The Rocks) samples and all of the Bermuda samples were dissolved in 4 N HCl to remove the CO_2 first for ^{14}C analyses (DRUFFEL, 1989).

The Sr extraction and purification chemistry procedures used were reported by BOWEN (1970) and LIVINGSTON *et al.* (1974). As ^{90}Y (daughter of ^{90}Sr) has a much shorter half-life of 64 hours and a maximum beta energy of 2.27 MeV, the ^{90}Y activity was determined using anticoincidence gas proportional beta counters (NOSHKIN and DEAGAZIO, 1966) and the ^{90}Sr activity derived from this, correcting for any Sr loss by analyzing for stable Sr carrier using Atomic Absorption Spectrophotometry. Uncertainties reported here are related to a least square fitting of the data to a 64 hour decay curve assuming a 70% confidence interval.

All ^{90}Sr values are decay corrected to the year of accretion and reported as dpm/100 gm of coral which is essentially equivalent to dpm/100 L of seawater. One hundred grams of coral contains 1.00 mole of Ca and a Sr/Ca ratio of approximately 0.009. One hundred liters of seawater contains 1.06 moles of Ca, and a Sr/Ca ratio equal to 0.01. The $\text{Sr}/\text{Ca}_{\text{coral}}/\text{Sr}/\text{Ca}_{\text{seawater}}$ discrimination factor is between 1.03 to 1.08 for a temperature range of 25–30°C (SMITH *et al.*, 1979). Thus, this 3–8% enrichment allows for direct comparison of ^{90}Sr activities in corals and seawater. TOGGWEILER (1983, p. 209) concluded that ^{90}Sr concentrations in corals were valid recorders of seawater ^{90}Sr concentration by comparing WeatherShip E ^{90}Sr seawater data near Bermuda with his Bermuda coral record.

Pu was extracted and analyzed in eight of "The Rocks" Florida samples. A ^{242}Pu spike was added to the acidified coral sample as a yield monitor. Sr was first precipitated with concentrated HNO_3 and removed from the coral solutions, then Pu was removed by ion exchange and analyzed using the Pu electroplating process reported in

LIVINGSTON *et al.* (1975). The $^{239,240}\text{Pu}$ activity was measured in alpha counters previously described in MANN *et al.* (1975). A $\text{Pu}/\text{Ca}_{\text{coral}}/\text{Pu}/\text{Ca}_{\text{seawater}}$ discrimination factor of 1.8 ± 1.2 has been previously reported by BENNINGER and DODGE (1986) indicating an enrichment in coral.

RESULTS AND DISCUSSION

Strontium-90 measurements of the Florida Keys and Bermuda corals are listed in Table 1 and shown in Fig. 2. A striking feature of the Florida (The Rocks) ^{90}Sr curve (Fig. 2b) is the similarity to the general shape of the New York City (40°N) ^{90}Sr atmospheric deposition (surface air sampling, HASL, 1977; TOONKEL, 1980; Fig. 2a). The highest ^{90}Sr values are found for the 1964 growth band (August 1963–July 1964), slightly later than the maximum observed in the atmospheric deposition record (1963). TOGGWEILER and TRUMBORE (1985) and MADAZAR *et al.* (1987) have both reported similar shaped curves of ^{90}Sr in Oahu and Ft. Lauderdale corals, respectively, and BENNINGER and DODGE (1986) in a St. Croix $^{239,240}\text{Pu}$ coral record. The reduction of ^{90}Sr activity in the atmosphere during the '59 to '61 period (Fig. 2a) is recorded in the coral as a minimum in the '60 to '62 period (Fig. 2b). This 0.5–1.0 year lag time appears to

FIG. 2. (a) Fallout ^{90}Sr activities in New York City. The fallout data (HASL, 1977; TOONKEL, 1980) has been adjusted to represent a Florida coral year. (b) Sr-90 activities in *Montastrea annularis* from The Rocks, Florida Keys and *Diploria strigosa* from North Rock, Bermuda. Results from the second Florida Keys site, New Ground (see Fig. 1) are available for three years (see text). The temporal position of each point is fixed at the centroid of a coral year; for Florida, a coral year is August to July, but for Bermuda it is April to March. Vertical bars represent uncertainty in activity. The Sr-90 activity coral record in the Florida Straits is 30–120% higher during the peak fallout years than that for Bermuda.

represent the response time for ^{90}Sr in the atmosphere to be deposited in the water and incorporated in the skeletons of shallow ocean corals.

The general decrease of ^{90}Sr levels throughout the 1970s in the Florida coral is more gradual than the atmospheric deposition curve. This is expected, due to the longer residence time of ^{90}Sr in surface seawater than in the atmosphere. Sr-90 is removed quickly from the atmosphere by precipitation, whereas it remains dissolved in seawater and is removed only by mixing with subsurface waters, horizontal advection and by radioactive decay. Dissolved ^{90}Sr in runoff from continents over a period of several years also adds ^{90}Sr activity to surface seawater, prolonging this isotope's apparent residence time.

The ^{90}Sr results from Bermuda (North Rock) are listed in Table 1 and plotted in Figs. 2b and 3. The Bermuda coral was analyzed to obtain an annual central gyre record (Fig. 2b), as well as to compare our values with previously reported results from Bermuda (TOGGWEILER, 1983, Fig. 3). Direct comparisons are difficult, since one year bands were used for this study, while TOGGWEILER (1983) uses two year bands which smooth the overall curve particularly at the peak fallout years (1958–1960 and 1962–1966). The Bermuda results from TOGGWEILER (1983) represent a complete record, while our record only covers eight selected years between 1951 and 1982. TOGGWEILER (1983) verified his results by comparing his coral ^{90}Sr activities to surface seawater ^{90}Sr activities at Weathership "E" station (35°N, 48°W) which represents a location in the center of the North Atlantic gyre. We have verified our results by comparing our coral data to surface ^{90}Sr seawater data for a 10° by 10° area encompassing Bermuda (28–38°N, 60–70°W). The seawater data presented in

Table 2: Surface seawater ^{90}Sr and $^{239,240}\text{Pu}$ Activities in the Atlantic Ocean within a box 10–20°N, 50–75°W taken between 1970–1975. This data represents surface ^{90}Sr activities for the Caribbean Sea. $K_{\text{Sr}} = 1.040 \pm 0.008$ and $K_{\text{Pu}} = 1.8 \pm 1.2$ (BENNINGER and DODGE, 1986).

Date	Cruise	Median* ^{90}Sr	No. of samples	Median* $^{239,240}\text{Pu}$	No. of samples
Feb-March 1970	AII 56	13	10	0.12	3
March 1972	Knorr 25	13	18	0.115	4
Nov. 1973	AII 78	11.5	33	0.07	7
Feb. 1974	Knorr 37	14	6		
Feb. 1975	AII 86	13	11	0.12	4

* ^{90}Sr and $^{239,240}\text{Pu}$ in dpm/100 L seawater

Fig. 3 represents 14 surface ^{90}Sr activities taken during six cruises from 1966 to 1970 for the Bermuda area (H. D. LIVINGSTON, unpublished data). The two Bermuda coral data sets differ by approximately 50% following the peak fallout years. But the seawater data, presented to verify the coral results, falls between the coral ^{90}Sr curves. Although the seawater data matches our coral data better in 1970, we do not have enough data points throughout the 1965 to 1970 period to further confirm the closer fit of our coral data to the surface seawater data.

The major difference between the Florida and Bermuda ^{90}Sr coral activities is the absolute concentration (Table 1, Fig. 2). During the years of peak atmospheric delivery (1959–1965), the ^{90}Sr activity is 30–120% higher at the Florida coral site than at the Bermuda site. However, based on data of fallout *versus* latitude, it is expected that the Florida site would receive 10% less ^{90}Sr fallout than Bermuda (JOSEPH *et al.*, 1971).

Furthermore, ^{90}Sr activities in Florida corals are 1.3 to 2.8 times higher than surface seawater activities in the Caribbean Sea for 1970 to 1975 (H. D. LIVINGSTON, unpublished data). SARMIENTO and GWINN (1986) predicted an increase of only 1.1 for fallout ^{90}Sr in the surface waters of Florida compared to the Caribbean Sea. Table 2 lists Caribbean ^{90}Sr activities in the surface seawater taken from the location 10°–20°N, 50°–75°W. Based on the above considerations, the Florida coral ^{90}Sr record reflects an enrichment of this isotope relative to both the Caribbean surface water and the Bermuda coral. But the enrichment in the Florida coral occurs only during the peak fallout years. During the 1970s, ^{90}Sr fallout was minimal; thus, ^{90}Sr levels in the surface ocean during this period were influenced primarily by circulation effects. Upwelling waters derived from the equatorial Atlantic and transported *via* the Gulf Stream would lower the surface water ^{90}Sr levels. These waters might account for the lowering of the Florida coral ^{90}Sr activity relative to Bermuda by 1974.

A confirmation of the very high activity in the 1964 Florida coral band (100 dpm/100 g; Fig. 2b) comes from the duplication of the ^{90}Sr activity in the 1964 band recorded in the New Ground coral (97 dpm/100 g) which is separated by 100 miles from The Rocks location in the Florida Keys (Fig. 1, Table 1). Both corals appear to be influenced by the same ^{90}Sr -enriched water mass. The 1974 ^{90}Sr value for the New

FIG. 3. Sr-90 activities in *Diploria strigosa* from North Rock, Bermuda. The two sites include: (1) NNE of The North Rock, Bermuda approximately 15 km north of Bermuda at 10 m depth (this study). (These results are also shown in Fig. 2.) (2) North Rock, Bermuda approximately 15–25 km north of Bermuda as reported in TOGGWEILER (1983). Bermuda results reported by TOGGWEILER (1983) represent two years of coral growth as compared to one year of coral growth for the Bermuda results (this study) which has the effect of smoothing the ^{90}Sr activity curve particularly during the peak fallout years. Darkened circles represent surface seawater ^{90}Sr activities (dpm/100 L seawater which are interchangeable with coral units) taken for an area 28–38°N, 60–70°W which includes the Bermuda site (unpublished data, see text). One darkened circle in 1970 does not appear due to overlap with other data points. Vertical bars represent uncertainty in activity.

Table 3: Comparison of $^{239,240}\text{Pu}$ Activities for *Montastrea annularis* from The Rocks reef, Florida Keys and St. Croix, U.S.V.I. (BENNINGER and DODGE, 1986).

Year	$^{239,240}\text{Pu}$ Florida (dpm/kg)	$^{239,240}\text{Pu}$ St. Croix (dpm/kg)	$\text{Pu}_{\text{Florida}}/\text{Pu}_{\text{St. Croix}}$	$^{239,240}\text{Pu}/^{90}\text{Sr}$ The Rocks, Florida
1940	0.007±0.014			
1957	0.77±0.18	2.85±0.18	0.27	$(3.4\pm0.8) \times 10^{-3}$
1961	0.85±0.21	2.75±0.18	0.31	$(3.9\pm1.0) \times 10^{-3}$
1962	0.85±0.15	4.08±0.23	0.21	$(2.7\pm0.5) \times 10^{-3}$
1963	1.86±0.25	7.61±0.40	0.24	$(3.4\pm0.5) \times 10^{-3}$
1964	3.68±0.29	10.06±0.44	0.37	$(3.7\pm0.3) \times 10^{-3}$
1966	0.47±0.08	4.90±0.14	0.10	$(0.8\pm0.1) \times 10^{-3}$
1967	0.87±0.16	3.39±0.23	0.26	$(1.8\pm0.3) \times 10^{-3}$

Ground coral is 30% higher than The Rocks 1974 sample. Perhaps the New Ground location reflects a coastal input which is then diluted downstream at The Rocks location due to coastal circulation complexities (see Coastal Circulation section.)

Enrichment of ^{90}Sr along coastal regions and in river runoff was reported in BOWEN *et al.* (1974). The computed $^{137}\text{Cs}/^{90}\text{Sr}$ mean fallout ratio is 1.45 (HARLEY *et al.*, 1965; KUPFERMAN and LIVINGSTON, 1979) and remains essentially unchanged in the ocean (BOWEN *et al.*, 1974, 1980). But near-shore $^{137}\text{Cs}/^{90}\text{Sr}$ ratios equal 1.0. This was accounted for by the combination of enriched ^{90}Sr from river runoff and groundwater discharge and particle removal of ^{137}Cs due to high concentrations of particles along coastal regions (BOWEN *et al.*, 1974).

Pu -239,240 results in the Florida coral are listed in Table 3 and shown in Fig. 4. The Florida coral $^{239,240}\text{Pu}$ activity levels, like ^{90}Sr , reflect a direct response to the atmospheric fallout changes. Similar to the ^{90}Sr activity record, the peak $^{239,240}\text{Pu}$ activity year occurs in 1964, about one year after the atmospheric maxima. The 1966 Pu sample in Table 3 appears to be anomalously low. It is not clear whether this resulted from unknown analytical error or is a real effect due to coastal circulation effects and the position of the Loop Current during that year (see Oceanographic Considerations section below.)

Our $^{239,240}\text{Pu}$ data for The Rocks Florida coral are strikingly different from those reported by BENNINGER and DODGE (1986) for St. Croix corals (Fig. 4, Table 3). Though the shapes of the two records are similar, the Florida coral contains approximately 20–35% of the $^{239,240}\text{Pu}$ levels in the St. Croix corals, comparing six individual year results. (The 1966 result has not been included in this comparison.)

The $^{239,240}\text{Pu}/^{90}\text{Sr}$ ratios for The Rocks Florida coral are reported in Table 3. This ratio has two maxima of 0.0039 in 1961 and 0.0037 in 1964 which reflect the fresh fallout increases seen in the atmospheric record before the nuclear weapons test ban in 1962. This ratio in fresh fallout is 0.018 (HARLEY, 1975; SHOLKOVITZ, 1983). Following the 1964 maximum, this ratio decreases significantly with time. The decrease in the ratio illustrates that less fallout $^{239,240}\text{Pu}$ is available to the corals relative to fallout ^{90}Sr as time progresses, because less of the $^{239,240}\text{Pu}$ remains in the water column relative to ^{90}Sr . Sr is a conservative element and therefore

remains in the dissolved form, whereas Pu is more particle reactive and is scavenged from the water column. Pu removal is particularly pronounced in coastal areas where high concentrations of particles exist in the water column (SANTSCHI *et al.*, 1980; SHOLKOVITZ and MANN, 1987). Relative to Pu delivered to the surface land or sea, only 3% of Pu is removed from the surface water column in open ocean, whereas 90% is removed along coastal regions and 97% in lakes (SHOLKOVITZ, 1983). Consequently, the $^{239,240}\text{Pu}$ in the surface ocean nearer the continents is removed quicker due to the combined effects of dilution and scavenging from the water column as compared to the ^{90}Sr radionuclide which must be decreased by dilution and decay only.

OCEANOGRAPHIC CONSIDERATIONS

To interpret these ^{90}Sr activity levels in relation to ocean circulation and mixing processes, we consider the differences in vertical mixing for the Caribbean Current vs. the Atlantic Ocean. Changes in the Loop Current position (see Fig. 1) within the Gulf of Mexico may explain differences in the ^{90}Sr seasonal record in the Florida Keys coral.

Ocean water entering the Caribbean Sea/Yucatan Straits region is stratified due to subtropical temperature and salinity effects (WUST, 1964). The 1970s ^3H profiles east of the Caribbean at 12°N show strong subsurface maxima at 100–200 m, whereas Atlantic Ocean profiles at 30°N reflect small or no subsurface maxima (OSTLUND, 1984; OSTLUND *et al.*, 1976). BOWEN *et al.* (1972) discussed a subsurface maxima in ^{90}Sr at about 100 m in the Caribbean Sea and related this to mixing and circulation of shallow water masses in this region. Consequently, Caribbean/Yucatan Straits/Gulf Stream surface waters are more likely to retain and concentrate fresh fallout and continental runoff in the upper 100–200 m of the water column.

Bomb radiocarbon results provide further evidence of less communication between surface and subsurface waters in

FIG. 4. Comparison of $^{239,240}\text{Pu}$ activities in *Montastrea annularis* from The Rocks reef, Florida Keys and St. Croix, U.S.V.I. as reported by BENNINGER and DODGE (1986). Vertical bars represent uncertainty in activity value. The $^{239,240}\text{Pu}$ activities are significantly lower in the Florida coral as compared to the corresponding years in the Caribbean coral. (See Table 3.) This is due to the geochemistry of Pu in coastal waters (adsorption on particulates) as compared to the oceanic waters influencing the Caribbean coral.

the Florida Straits region. DRUFFEL (1989) studied upper ocean ventilation in the Sargasso Sea gyre using high precision bomb $\Delta^{14}\text{C}$ measurements. An increase in $\Delta^{14}\text{C}$ occurred earlier in Florida corals than in Bermuda corals. This is not due to coastal influences. Radiocarbon is introduced to the surface ocean *via* isotopic gas exchange, and with a relatively long turnover time (ten years, DRUFFEL and LINICK, 1978). Modelling the coral results indicates that the bomb ^{14}C signal was damped at Bermuda during 1960–1970 due to increased mixing with ^{14}C -poor subsurface waters (200–400 m) during 18° or more water formation that occurs during late winter (WORTHINGTON, 1976). Conversely, the thermocline in the Florida Straits during the winter months deepens to only 50–75 m, thus allowing a much smaller volume into which the bomb ^{14}C can be mixed, resulting in a higher concentration of this isotope in surface waters overall.

Seasonal ^{90}Sr activity levels in Florida corals are shown in Fig. 5b and listed in Table 4. Both 1973 and 1974 show the lowest activity during the winter months of January to March. Since it has been shown that temperature affects the Sr/Ca ratio in corals to only a minor extent (less than 10% change over 12°C range reported in SMITH *et al.*, 1979) or not at all (GOREAU, 1977; SWART, 1981), the seasonal variation in ^{90}Sr activity in corals is likely due to a change in the source of water to the coral site. The 30% lower coral ^{90}Sr values during the winter could not be due to increased depth of the mixed

Table 4: ^{90}Sr activities measured in four separate seasons as indicated for 1973 and 1974 in *Montastrea annularis* at The Rocks reef in the Florida Keys.

Season	1973 Activity*	1974 Activity*
Jan–March	16.8±3.6	13.4±1.7
April–June	20.7±1.9	17.3±1.4
July–Sept	24.8±0.7	20.4±0.5
Oct–Dec	21.2±0.9	20.3±0.5

* ^{90}Sr dpm/100 g coral

layer and entrainment of lower ^{90}Sr waters, since ^{90}Sr levels in the subsurface waters are the same or higher than those in surface waters (BOWEN *et al.*, 1972, 1974; LIVINGSTON *et al.*, 1985). There is preliminary evidence that this seasonal variation in ^{90}Sr activity in corals is related to the position of the Loop Current.

Waters from the North Equatorial Current and the Guiana Current primarily pass through the Lesser Antilles into the Caribbean Sea, with eventual passage through the Yucatan Channel (WUST, 1964) into the Gulf of Mexico. During some periods of the year this water travels nearly directly north toward the Florida Keys, then turns east into the Florida Straits. However, the degree of penetration of this current into the Gulf of Mexico typically increases with time, forming a progressively larger anticyclonic Loop Current into the Gulf (Fig. 1) before exiting into the Florida Straits (MAUL, 1977; LEIPPER, 1970; HUH *et al.*, 1981; VUKOVICH *et al.*, 1979; MOLINARI and MAYER, 1982; MOLINARI *et al.*, 1977; NOWLIN and McLELLAN, 1967; BEHRINGER *et al.*, 1977). An annual cycle of Loop Current growth and eventual decay due to eddy separation has been documented over various periods (BEHRINGER *et al.*, 1977; STURGES and EVANS, 1983; see Fig. 5a). It has been shown that this Loop Current gyre in the Gulf has extended almost to the Mississippi Delta in August 1973 (MAUL, 1977) and onto the West Florida Continental shelf passing within 8 km of shore in February 1977 (HUH *et al.*, 1981). These extensive intrusions mix with and entrain north and east coastal Gulf waters significantly changing the properties (*i.e.* salinity, temperature, etc.; MAUL, 1977; MOLINARI and MAYER, 1982) of the water mass exiting cyclonically into the Florida Straits. For example, very low salinity water (24‰) observed all along the edge of the Florida Current off the Florida Keys coincided with the August 1973 maximum intrusion event (MAUL, 1977).

Figure 5a represents the Loop Current position time series for 1973 and 1974 taken from STURGES and EVANS (1983). When the maximum intrusion occurred in these years, the Florida coral ^{90}Sr activities coincide with seasonally high values (Fig. 5b). We suspect that the coral registers more ^{90}Sr during the summer months, possibly due to increased entrainment of coastal water as a result of northward Loop Current intrusion into the Gulf of Mexico. It is difficult to resolve the time lag between the ^{90}Sr activity increase in the coral as a response to the Loop Current penetration into the Gulf, since crucial position data is not available for November and December, 1973. Additionally, the resolution in the coral

FIG. 5. (a) Position of northern edge of the Loop Current based on available hydrographic data for 1973 and 1974 (STURGES and EVANS, 1983). No data points are available for months 11, 12/73 and 7, 8, 9, 12/74. A complete time series (1965 to 1978) assigning monthly points for the Loop Current position based on a cubic spline fit through the available hydrographic data appears in STURGES and EVANS (1983). (b) Seasonal ^{90}Sr activities for 1973 and 1974 in *Montastrea annularis* from The Rocks reef, Florida Keys. Three months of coral growth were sectioned from the core for ^{90}Sr analysis for two consecutive years. Vertical bars represent uncertainty in activity value. See Table 4 for comparison of actual values.

data is in three month intervals, whereas it is in one month intervals for the Loop Current data.

An extensive and prolonged Loop Current intrusion occurred in 1966 (STURGES and EVANS, 1983). This major intrusion event might account for our very low 1966 coral Pu value due to particle laden coastal water influence in the Gulf Stream which would remove Pu from the water column.

COASTAL CIRCULATION NEAR THE FLORIDA KEYS

To investigate the possibility of coastal influence on the ^{90}Sr coral record at The Rocks reef in greater detail, it is necessary to examine the circulation patterns in the Florida Straits and around the Florida Keys. Extensive meanders of the Gulf Stream through the Florida Straits create localized circulation patterns off the Florida Keys and southeastern Florida coast important to this study. Currents in the axis of the Gulf Stream reach speeds up to 200 cm/sec. However, cyclonic spin-off eddies, caused by offshore meanders of the stream, can occur all along the north and western edge of the Florida Current causing current reversals along the adjacent coastal regions (LEE and MAYER, 1977; LEE and MOOERS, 1977; LEE, 1975; BROOKS and NILER, 1975; see Fig. 6). These cyclonic eddies continually exchange Florida Current water with coastal water on the shelf. The Pourtales Terrace is the extensive 30–50 km platform (<200 m depth) off the lower and middle Florida Keys (Fig. 6a). It is characterized by persistent coastal and bottom countercurrents and longer duration cyclonic gyres than the weekly produced inshore cyclonic gyres created downstream off Miami (Fig. 6b; LEE, 1975; BROOKS and NILER, 1975). These persistent cyclonic flows provide a mechanism for fish larvae transport from remote upstream spawning grounds (*i.e.* north of Key West and inside Florida Bay) to downstream coastal and nearshore nursery areas such as inside the upper Keys (T. N. LEE, pers. commun., REHRER *et al.*, 1967; MURPHY *et al.*, 1975). Additionally, the Gulf Stream is in a curving mode off the Florida Keys and is therefore located more offshore compared to its location along the coast off the narrow Miami Terrace. The Gulf Stream runs straight all along the lower eastern Florida coast. Therefore, corals along the eastern coast of Florida are expected to be more directly influenced by Gulf Stream water than those along the southern coast off the Keys.

BOYLE *et al.* (1984) showed that copper enrichments in the surface waters of the Florida Current could be accounted for from river sources to the Gulf Stream. However, we have concluded that the ^{90}Sr in the Mississippi River runoff could not account alone for our ^{90}Sr Florida coral enrichments if integrated throughout the upper 50 m of Gulf transport. SIMPSON *et al.* (1987) reports an estimated ^{90}Sr activity of 1.79 pCi/L in the Hudson River for 1964, whereas MENZEL (1974) reports an average ^{90}Sr activity of 2.08 pCi/L in U.S. streams for the same year. Using similar assumptions made by BOYLE *et al.* (1984) of $10 \cdot 10^6 \text{ m}^3/\text{sec}$ flow for the upper 100 m of the Florida Current and of $0.01 \cdot 10^6 \text{ m}^3/\text{sec}$ for the Mississippi River flux (SHILLER and BOYLE, 1987), a ^{90}Sr activity of 444 dpm/100 L coming from the Mississippi River would be a minimal signal diluted rapidly when mixed with

FIG. 6. Cyclonic spin-off eddies shown over (a) the Pourtales Terrace (T. N. LEE, pers. commun.) and (b, enlarged inset) the Miami Terrace. (adapted from LEE and MAYER, 1977).

Gulf Stream waters carrying a surface fallout ^{90}Sr activity of approximately 50 dpm/100 L for 1964. However, as the axis of the Gulf Stream is held offshore due to meanders and the Pourtales Terrace, coastal eddies carrying constantly exchanged waters from the coastal shelf and the edge of the Gulf Stream might concentrate coastal waters over the Florida Keys corals.

Closer range influences must address Florida Bay runoff effects. LANDSAT images and ship track records in a Loop Current study (MAUL, 1977) and digital thermal infrared data (ROBERTS *et al.*, 1982) record evidence of Florida Bay runoff advecting through the Keys. In fact, winter runoff water passing from the Bay through the Keys producing lethally cold ($<12^{\circ}\text{C}$) shallow-water accounts for the absence of a living reef tract across tidal passes within the Keys (ROBERTS *et al.*, 1982). The Florida reef tract is generally protected by the buffering effect of warm oceanic water.

HUDSON (1981) transplanted *Montastrea annularis* along an inshore-offshore transect from Snake Creek to Crocker Reef to study the effects of environmental stress on reef-building corals. He attributed the death or reduced growth of corals closest to the tidal pass at Snake Creek to water coming from the Florida Bay which can have temperatures outside the tolerance range of $13.9\text{--}32^{\circ}\text{C}$ for *Montastrea annularis*. Since our coral (The Rocks) is located 4 km from Snake Creek in 4 m of water, it is possible that it is influenced by runoff from the Florida Bay. We believe the combination of close range runoff and coastal eddies could concentrate the ^{90}Sr activities in the shallow waters off the Florida Keys, much like a blender that recirculates fluid within a finite volume.

CONCLUSION

The time histories of atmospheric introduction and fallout of ^{90}Sr and $^{239,240}\text{Pu}$ are clearly reflected in a coral from the Florida Keys site (The Rocks). The coral activity curve for these isotopes reflects a 0.5 to 1.0 year lag relative to the atmospheric activity curve. The ^{90}Sr activity reflects a more gradual reduction in the 1970s in coral relative to the atmosphere, due to longer residence time of Sr in the surface ocean than in the atmosphere.

The Florida coral record documents enrichment of ^{90}Sr and depletion of $^{239,240}\text{Pu}$ compared to fallout predictions and previous coral records of these isotopes from central gyre localities. These two and three fold differences in isotope activities in the coral cannot be accounted for by changes in temperature and chemical composition of the seawater or differences in physical parameters of the coral such as growth or coral species. We conclude that the enrichment of ^{90}Sr in the Florida Keys coral record is likely the result of continental runoff which has extended exposure to the coral due to coastal circulation patterns in this region. The concomitant depletion of $^{239,240}\text{Pu}$ is due to removal from the water column by particles in coastal waters.

There appears to be evidence that the ^{90}Sr and $^{239,240}\text{Pu}$ activities in corals along coastal regions may be strongly influenced not only by geochemistry, but by circulation effects. Even two Florida Keys corals, although they had similar ^{90}Sr levels in 1964, differed by 30% in 1974. The oceanic inputs of ^{90}Sr and $^{239,240}\text{Pu}$ from fallout and runoff ceased by the early 1970s. Ocean and coastal circulation patterns became the dominant factor influencing these activities in corals. This suggests that site specific considerations must be addressed before coral ^{90}Sr records can be interpreted. These considerations might be viewed as limitations to the use of coral

^{90}Sr records for large scale circulation information. However, coastal coral ^{90}Sr records may become a very useful tool for interpreting local circulation patterns.

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