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Cuban coral traces annual hydrologically driven variability in δ^{234}U values since the end of the Little Ice Age

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Mississippi Mass calculation

Considering the present-day strong seasonal variability in the U concentration and isotopic composition of the Mississippi River (Grzymko et al., 2007), it cannot be assumed that the uranium concentration and $^{234}U/^{238}U$ activity ratio have behaved conservatively in the Mississippi River over the last 240 years. Binary isotope mixing according to Equation (S1) (Roy-Barman et al., 2016) was applied to estimate the potential fraction of the Mississippi River-derived U isotopes in the Gulf of Mexico throughflow.

$$f_r = \frac{\delta_c * C_c - \delta_{sw} * C_{sw}}{\delta_r * C_r - \delta_{sw} * C_{sw}} \tag{S1}$$

Here, δ_C is the coral $\delta^{234}U$ ratio and C_C is the coral U concentration converted into the corresponding seawater concentration (assuming 3.5 µg/g - 3.3 µg/l at 35,9 PSU). Any other influences on the U concentration, such as temperature, carbonate ion concentration and the coral growth rate, are neglected. Similarly, δ_{SW} is the open ocean seawater $\delta^{234}U$ ratio, and C_{SW} is its U concentration (presumed to be constant, $[U]_{Sw} >> [U]_{river}$). Finally, δ_r is the Mississippi $\delta^{234}U$ ratio range, and C_r is the U concentration range. For this two end-member isotope system, the past seawater U concentration remains unknown but is likely to vary within small ranges of a few percent from modern values. To overcome this difficulty, we here used the mean coral U concentration (2.27 µg/l) as a representative of seawater. Taking these limitations into account, the proportion of Mississippi River freshwater (f_r) reaching the coral site during the LIA, where the highest $\delta^{234}U$ ratios are recorded, is estimated to vary between 0% and 3.0%.

The seawater U concentration may vary independently of open ocean and freshwater mixing, due to evaporation, for example (Ivanovich & Harmon, 1992). Additionally, U incorporation during coral calcification may vary depending on the growth rate (Inoue et al., 2011), seawater pH (Inoue et al., 2011), carbonate ion concentration, dissolved inorganic carbon (DeCarlo et al., 2016; Patterson et al., 2021), and temperature (Ourbak et al., 2006; Patterson et al., 2021), causing subtle changes in the coral U concentration independent of the balance between seawater and freshwater. Consequently, any quantitative estimate will remain speculative without monitoring all the above aspects. Nonetheless, the Mississippi freshwater volume contribution to the Gulf of Mexico throughflow is currently less than a 0.5 ‰ in mass balance (Grzymko et al., 2007; Leaman et al., 1995), leading to subtle salt changes and minor changes in oxygen isotopes overprinted by net evaporation. Hence, we must assume that changes in the U concentration of seawater caused by the contribution of the Mississippi are minimal. Consequently, any observed variations in isotopic composition must reflect a major shift in the supply of U and/or an increase in the $\delta^{234}U$ value of river discharge.

In summary, we suggest that the deviation of the Cuban coral $\delta^{234}U$ values from a constant open ocean $\delta^{234}U$ value is most likely driven by two independent excess U sources. First, the local runoff coupled with the variance in precipitation on the island of Cuba likely resulted in minor reductions in the U isotope values compared with the open ocean mean values. The observed correlation between a decrease in isotopic composition and an increase in precipitation accounts for approximately 50% of the variability in the $\delta^{234}U$ time series from 1850–present. At the end of the LIA period (1778-1846), when the climate cooled in the Northern Hemisphere and the tropical climate was unstable, a second process influenced the time variance of the coral $\delta^{234}U$ values. Most likely, the distant influence of the excess ^{234}U contribution from the Mississippi River into the strong persistent throughflow of the Caribbean current caused a moderate and punctuated strong increase in seawater and thus coral $\delta^{234}U$. Both local precipitation and distant freshwater runoff must influence the $\delta^{234}U$ time variance and mean of the coral record. Here, these two distinct contributions did not modify the long-term average coral $\delta^{234}U$ value, which is identical to the mean global ocean value. In addition, the variance in $\delta^{234}U$ is less than ±3 ‰ when the single elevated value in 1792 is excluded. This finding strongly constrains the expected variance in the initial $\delta^{234}U$ values of fossil corals within the Caribbean.

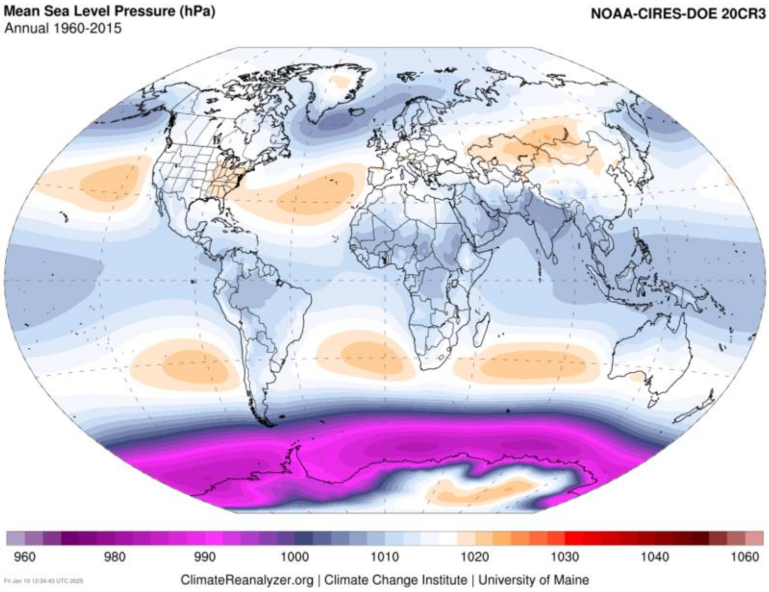

Figure S1: Mean annual sea level pressure in hPa spanning 1960 – 2015 using NOAA-Cires-Doe 20CR3. Cuba lies on the southwestern edge of the high-pressure system over the Atlantic and is influenced by the extension of this pressure system.

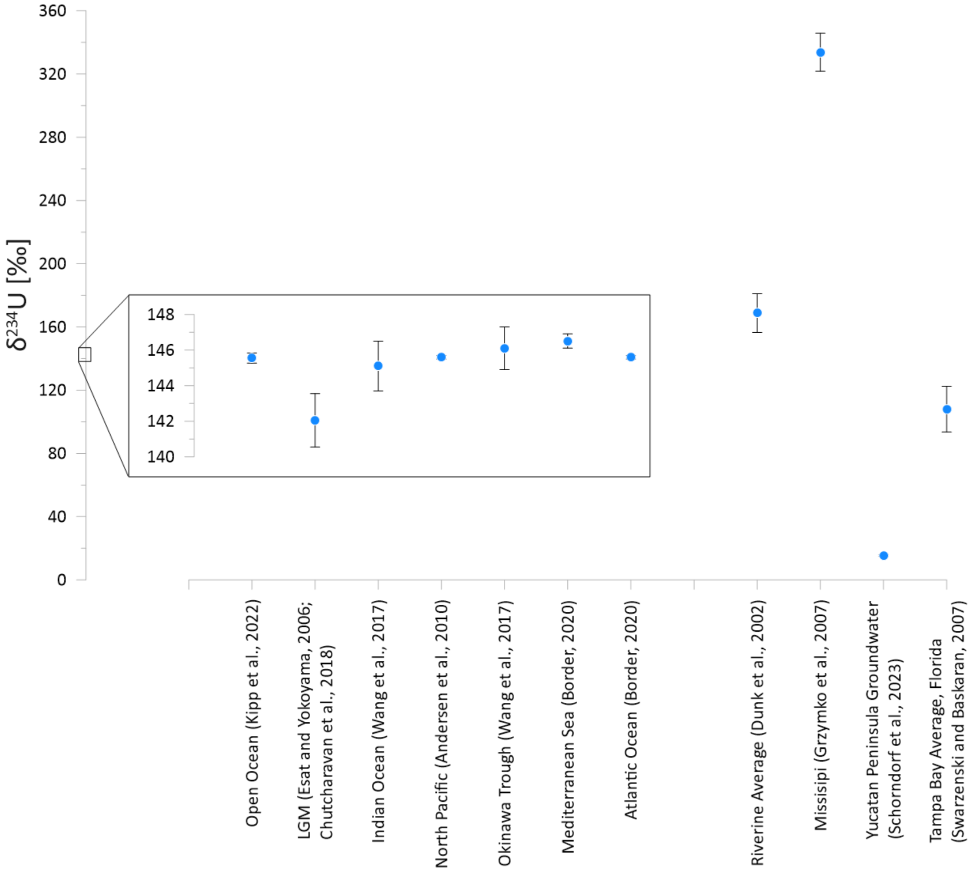

Figure S2. Global and regional δ^{234}U values compiled from modern marine, riverine, and groundwater sources. Marine values include open ocean, LGM reconstructions, and specific basins (e.g., Indian Ocean, Mediterranean Sea), showing δ^{234}U generally between ~142‰ and 147‰. Riverine and groundwater sources display broader variability, with particularly high values observed in the Mississippi River and Yucatán Peninsula groundwater. A close-up inset highlights the narrow range of marine δ^{234}U values relevant for comparison with proxy records.

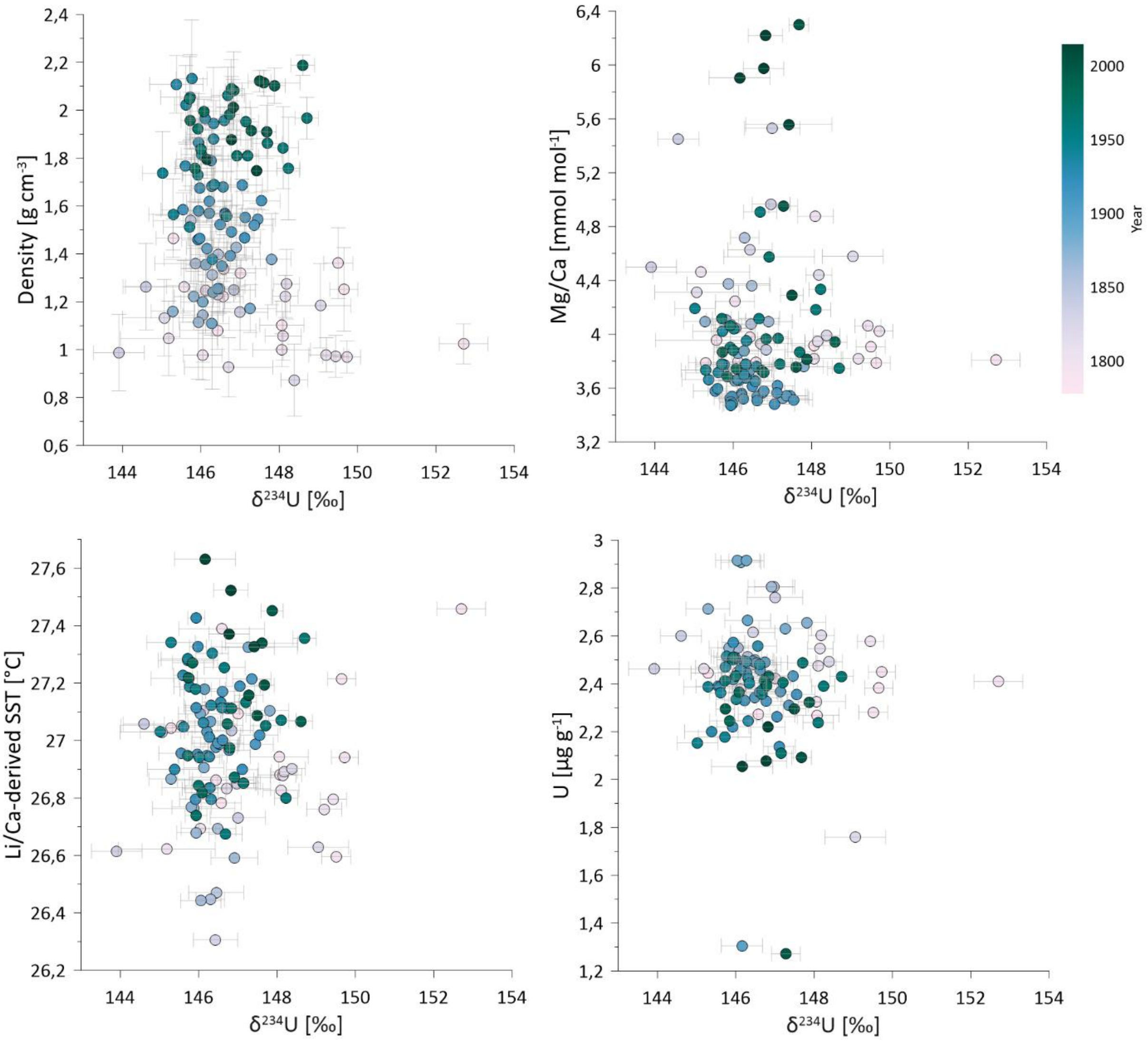

Figure S3. no correlation between the coral density, coral Mg/Ca, Li/Ca derived SST (Alonso-Hernández et al., 2022) or U concentration and δ^{234}U values is visible, implying no δ^{234}U signal variations due to vital effects or diagenesis. Colour scale denotes sample year.

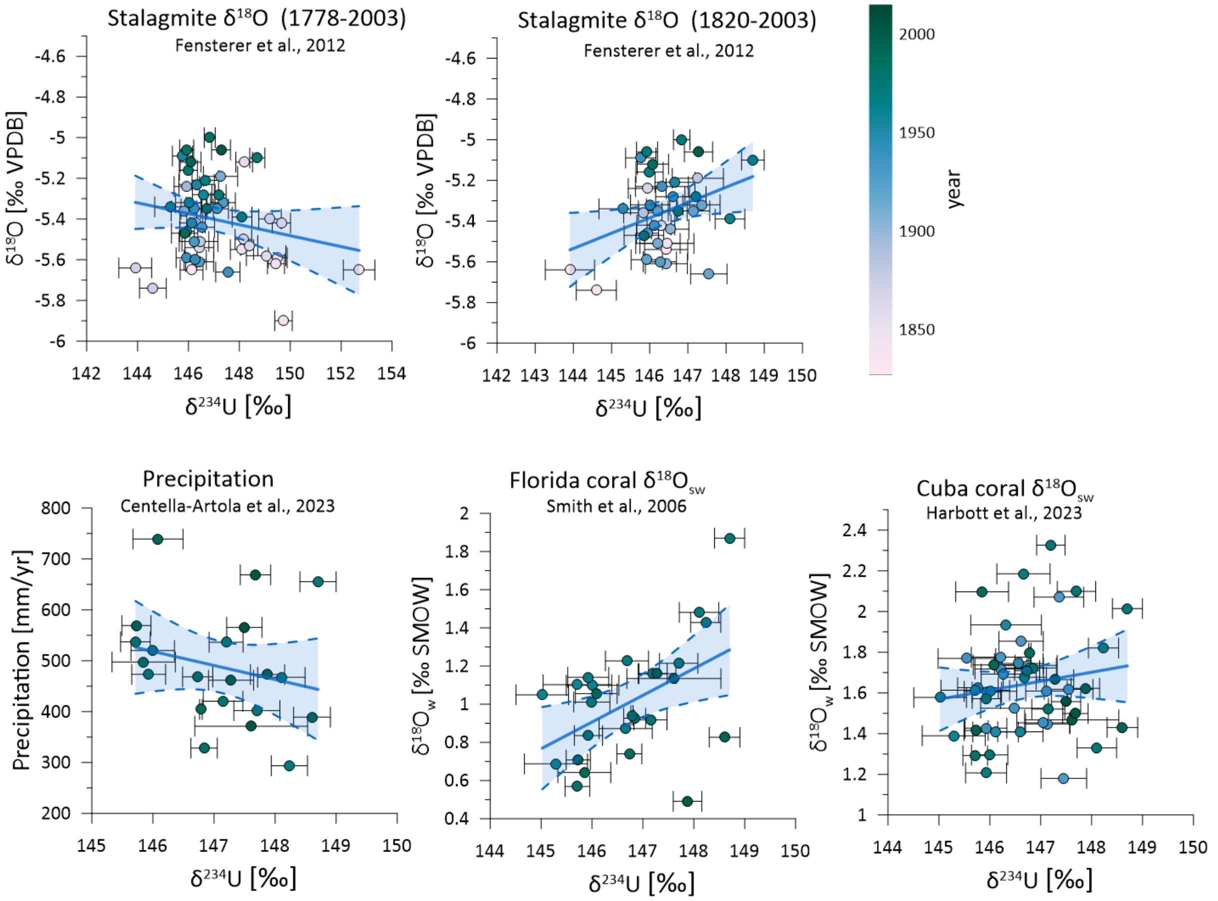

Figure S4. Correlations between δ^{234}U and regional hydroclimate proxies over the past two centuries. Top panels show δ^{18}O from stalagmites (Fensterer et al., 2012) plotted against δ^{234}U for the full (1778–2003) and post-1820 intervals. Bottom panels compare δ^{234}U with annual precipitation from the Cuban coast (Centella-Artola et al., 2023), and seawater δ^{18}O from Florida (Smith et al., 2006) and Cuba (Harbott et al., 2023) coral records. Shaded regions indicate 95% confidence intervals around linear regressions. Colour scale denotes sample year.

Table S1. δ^{234}U measurements together with concentrations of ^{238}U and ^{234}U of annually integrated CSM_1. δ^{234}U values are given normalised to HU1 = 0 and HU1 ≠ O after (Cheng et al., 2013). Measurement uncertainties are 2σ analytical errors

Lab. #	**Year**	**δ^{234}U (‰)**	**Error δ^{234}U (‰)**	**^{234}U (pg/g)**	**Error ^{234}U (pg/g)**	**^{238}U (µg/g)**	**Error ^{238}U (µg/g)**
12021	1777	146.02	0.39	152.59	0.44	2.42	9.91E-05
12037	1778	145.59	0.51	144.06	0.70	2.27	3.50E-04
12022	1780	147.09	0.57	143.30	0.61	2.27	1.25E-04
12038	1781	145.45	0.97	143.39	1.55	2.26	7.63E-04
12023	1783	145.14	0.45	151.19	0.35	2.38	1.18E-04
12039	1784	144.95	0.48	151.61	0.55	2.45	1.40E-04
12024	1786	148.67	0.36	151.17	0.40	2.38	8.88E-05
12040	1787	145.61	1.09	150.11	1.18	2.37	5.47E-03
12025	1789	148.53	0.37	145.86	0.35	2.28	8.13E-05
12041	1790	144.59	1.56	150.04	2.86	2.37	1.02E-03
12026	1792	151.72	0.62	152.70	0.53	2.41	1.40E-04
12042	1793	147.08	0.74	147.37	1.37	2.33	5.26E-04
12027	1795	148.75	0.34	154.70	0.40	2.45	1.13E-04
12043	1796	145.07	0.50	155.12	0.75	2.45	3.23E-04
12028	1798	148.46	0.33	162.69	0.48	2.58	1.05E-04

Lab. #	**Year**	**δ^{234}U (‰)**	**Error δ^{234}U (‰)**	**^{234}U (pg/g)**	**Error ^{234}U (pg/g)**	**^{238}U (µg/g)**	**Error ^{238}U (µg/g)**
12029	1801	147.12	0.46	158.76	0.74	2.48	1.38E-04
12045	1802	144.20	1.24	157.88	2.06	2.58	3.62E-04
12030	1804	148.22	0.44	154.93	0.47	2.46	1.22E-04
12047	1808	145.73	1.13	170.98	2.01	2.62	3.16E-04
12032	1810	147.18	0.33	159.03	0.37	2.55	9.93E-05
12033	1813	147.21	0.33	161.74	0.56	2.60	1.22E-04
12049	1814	144.09	1.07	163.12	1.83	2.59	3.46E-04
12034	1816	148.07	0.78	109.21	0.58	1.76	1.08E-04
12035	1819	147.40	0.34	156.45	0.46	2.49	9.98E-05
12085	1826	145.45	0.57	162.14	0.39	2.61	1.08E-04
12742	1830	144.57	0.50	162.89	0.74	2.65	1.95E-04
12086	1831	143.93	0.64	154.86	0.83	2.46	1.91E-04
12237	1833	146.02	0.71	174.42	0.21	2.76	6.55E-05
12743	1835	145.99	0.49	172.83	0.69	2.81	1.68E-04
12087	1836	143.62	0.52	160.43	0.66	2.60	1.79E-04
12238	1838	144.77	0.97	174.18	0.23	2.75	8.20E-05
12088	1841	145.86	0.56	152.50	0.88	2.43	1.66E-04
12089	1846	145.47	0.71	153.65	1.00	2.48	2.23E-04
12090	1851	145.31	0.36	156.02	0.36	2.51	1.02E-04
12091	1856	145.50	0.47	156.50	0.59	2.50	1.39E-04
12242	1858	144.89	1.05	173.50	0.39	2.74	9.78E-05
12092	1861	145.07	0.51	157.41	0.63	2.55	1.50E-04
12243	1863	145.93	0.60	178.40	0.34	2.81	1.15E-04
12093	1866	144.95	0.50	155.21	0.50	2.51	1.15E-04
12094	1871	144.84	0.50	158.06	0.68	2.55	1.53E-04
12245	1873	144.31	0.56	172.51	1.01	2.71	2.39E-04
12095	1876	146.83	0.49	164.82	0.62	2.66	1.37E-04
12246	1878	145.15	0.51	180.49	0.80	2.91	1.85E-04
12096	1881	146.28	0.67	161.91	0.71	2.63	1.95E-04
12247	1883	145.07	0.56	180.41	0.55	2.92	1.54E-04
12248	1888	145.29	0.45	181.79	0.73	2.91	1.75E-04
12098	1891	145.33	0.58	165.23	0.37	2.66	1.16E-04
12250	1898	145.17	0.52	108.21	0.27	1.31	9.38E-05
12009	1901	145.45	0.55	156.06	0.64	2.50	1.42E-04
12008	1903	145.56	0.49	154.28	0.59	2.49	1.16E-04
12007	1905	145.77	0.58	154.00	0.52	2.49	1.28E-04
12006	1907	146.47	0.45	151.55	0.53	2.43	1.12E-04
12005	1908	146.38	0.49	142.45	0.54	2.31	1.31E-04
12004	1909	145.63	0.44	153.61	0.63	2.46	1.46E-04
12003	1910	145.24	0.58	159.71	0.89	2.50	1.92E-04
12002	1911	145.79	0.53	147.50	0.61	2.33	1.36E-04
11850	1912	146.16	0.43	148.99	0.60	2.37	1.23E-04
12001	1913	145.51	0.67	148.97	1.10	2.34	2.00E-04
12000	1915	144.57	0.57	150.67	0.58	2.39	1.39E-04
11999	1916	145.24	0.51	147.05	0.56	2.33	1.16E-04
11851	1917	146.57	0.48	148.37	0.50	2.36	1.31E-04

Lab. #	**Year**	**δ^{234}U (‰)**	**Error δ^{234}U (‰)**	**^{234}U (pg/g)**	**Error ^{234}U (pg/g)**	**^{238}U (µg/g)**	**Error ^{238}U (µg/g)**
11998	1918	146.07	0.52	143.79	0.58	2.26	1.33E-04
11997	1919	144.94	0.41	140.69	0.64	2.22	1.31E-04
11996	1920	146.13	0.41	134.46	0.47	2.14	1.08E-04
11993	1921	144.99	0.30	154.46	0.15	2.50	5.36E-05
11852	1922	145.63	0.44	146.90	0.33	2.37	1.40E-04
11992	1923	144.96	0.29	160.20	0.27	2.57	7.70E-05
11991	1924	145.00	0.22	155.20	0.15	2.51	5.02E-05
11990	1925	145.28	0.43	151.19	0.28	2.44	7.55E-05
11853	1927	144.96	0.21	149.59	0.11	2.42	4.40E-05
11988	1928	144.62	0.24	147.59	0.18	2.39	5.36E-05
11987	1929	145.59	0.48	157.73	0.64	2.56	1.52E-04
11986	1930	144.40	0.68	138.11	0.75	2.20	1.76E-04
11985	1931	145.34	0.36	137.84	0.40	2.25	9.71E-05
11854	1932	145.13	0.45	152.45	0.55	2.46	1.23E-04
11984	1933	145.31	1.23	151.21	1.44	2.45	3.10E-04
11983	1935	145.29	0.32	153.59	0.36	2.49	9.10E-05
11982	1936	145.34	0.69	148.15	1.05	2.42	2.56E-04
11855	1937	144.75	0.75	154.36	1.27	2.47	2.37E-04
11981	1938	144.78	0.40	154.39	0.53	2.52	1.16E-04
11980	1939	144.64	0.29	145.61	0.26	2.36	6.79E-05
11979	1941	145.62	0.44	154.11	0.57	2.48	1.29E-04
11856	1942	145.36	0.41	147.62	0.45	2.40	1.14E-04
11978	1944	144.73	0.67	134.27	0.53	2.18	1.22E-04
11977	1945	144.31	0.62	150.82	0.80	2.39	1.50E-04
11976	1946	144.04	0.52	135.07	0.60	2.15	1.28E-04
11857	1947	145.03	0.52	145.84	0.71	2.33	1.29E-04
11970	1954	145.68	0.52	146.68	0.50	2.36	1.06E-04
11969	1955	144.95	0.40	155.47	0.43	2.50	1.09E-04
11859	1957	145.71	0.42	147.23	0.29	2.37	9.13E-05
11933	1959	147.25	0.30	152.74	0.31	2.39	7.79E-05
11932	1960	147.12	0.39	143.46	0.32	2.24	8.29E-05
11931	1963	147.72	0.30	168.75	0.36	2.43	7.95E-05
11930	1964	146.72	0.38	158.54	0.53	2.49	1.29E-04
11929	1965	146.22	0.28	151.70	0.25	2.41	7.32E-05
11861	1967	145.02	0.36	150.38	0.23	2.43	6.55E-05
11928	1968	146.17	0.32	133.64	0.23	2.11	6.96E-05
11927	1969	145.94	1.16	154.89	1.65	2.45	3.22E-04
11926	1970	144.73	0.24	152.46	0.20	2.41	6.25E-05
11925	1971	144.95	0.28	159.25	0.22	2.51	6.76E-05
11924	1973	145.86	0.22	153.26	0.15	2.43	5.39E-05
11923	1978	145.75	0.25	150.17	0.22	2.41	6.08E-05
11862	1981	145.81	0.02	147.22	0.63	2.39	1.38E-04
11864	1982	144.87	0.52	140.01	0.57	2.25	1.42E-04
11894	1985	145.10	0.41	146.10	0.35	2.36	9.83E-05
11893	1988	147.62	0.30	144.99	0.35	2.30	8.93E-05
11866	1992	144.75	0.24	142.36	0.18	2.29	5.82E-05

Lab. #	Year	$\delta^{234}U$ (‰)	Error $\delta^{234}U$ (‰)	^{234}U (pg/g)	Error ^{234}U (pg/g)	^{238}U (µg/g)	Error ^{238}U (µg/g)
11892	1994	146.90	0.28	145.25	0.38	2.32	1.07E-04
11867	1997	146.63	0.92	23.81	0.04	0.38	1.35E-05
11876	1998	146.30	0.37	79.18	0.12	1.27	3.73E-05
11875	1999	146.70	0.25	130.65	0.19	2.09	6.30E-05
11874	2003	146.52	0.29	143.38	0.41	2.30	1.13E-04
11873	2011	146.44	1.10	134.51	1.58	2.14	2.99E-04
11870	2012	145.18	0.78	129.87	0.77	2.05	1.74E-04
11872	2013	145.84	0.44	138.60	0.47	2.22	9.95E-05
11871	2014	145.79	0.52	130.13	0.52	2.08	1.14E-04

Table S2. $\delta^{18}O_w$ of a coral from the Florida Keys (Smith et al., 2006) was calculated with the SST calculated from Sr/Ca ratios by using the equation $Sr/_{Ca} = -0.0282 * SST + 9.962$ (Smith et al., 2006) with this $SST_{Sr/Ca}$, the $\delta^{18}O_w$ is calculated from the measured $\delta^{18}O_C$ by $\delta^{18}O_W = ((SST_{Sr/_{Ca}} - 5.33)/4.519) + \delta^{18}O_c$ (Leder et al., 1996; Ren et al., 2003)

year	$SST_{Sr/Ca}$ (°C)	$\delta^{18}O_w$ (‰ SMOW)
2002	29.54	1.16
2001	28.17	1.13
2000	27.76	1.04
1999	27.38	0.95
1998	24.81	0.49
1997	26.81	0.84
1996	25.89	0.71
1995	27.70	0.98
1994	27.57	0.99
1993	25.87	0.66
1992	26.33	0.83
1991	27.44	0.97
1990	28.94	1.11
1989	28.44	1.06
1988	26.64	0.81
1987	24.24	0.35
1986	26.51	0.64
1985	27.78	0.94
1984	28.28	1.06
1983	26.61	0.75
1982	26.21	0.74
1981	28.36	1.14
1980	26.02	0.68
1979	26.88	0.92
1978	25.70	0.71
1977	26.13	0.93
1976	24.54	0.57
1975	26.26	0.84
1974	25.22	0.57
1973	26.81	0.68

year	$SST_{Sr/Ca}$ (°C)	$\delta^{18}O_w$ (‰ SMOW)
1972	27.02	0.92
1971	27.43	1.01
1970	27.22	0.99
1969	27.86	1.16
1968	28.03	1.21
1967	31.57	1.87
1966	29.80	1.49
1965	29.68	1.37
1964	28.91	1.48
1963	28.42	1.43
1962	28.79	1.47
1961	28.62	1.23
1960	29.20	1.40
1959	27.79	1.14
1958	26.76	0.87
1957	27.73	1.07
1956	29.22	1.42
1955	30.07	1.56
1954	30.32	1.52
1953	29.50	1.22
1952	28.52	1.21
1951	27.94	1.10
1950	27.89	1.05
1949	27.22	0.69
1948	29.25	1.10
1947	27.67	0.92

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