# Evidence for Preindustrial Variations in the Marine Surface Water Carbonate System from Coralline Sponges

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# **Evidence for Preindustrial Variations in the Marine Surface Water Carbonate** System from Coralline Sponges

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Abstract: Coralline sponge skeletons are excellent tools for reconstructing the carbon isotope history of dissolved inorganic carbon (DIC) in tropical surface waters. Carbon isotope records from coralline sponges clearly reflect the industrial <sup>12</sup>C increase in atmospheric CO<sub>2</sub> with a precision that permits quantitative interpretations. We find from a set of  $\delta^{13}$ C records of four Caribbean sponge specimens that the isotopic response of surface water DIC to the changing isotopic composition of atmospheric CO<sub>2</sub> varied dynamically during the last century, depending on the rate of atmospheric change. Three of our sponges provide 600-year long  $\delta^{13}$ C records. For the first time, we can reconstruct surface water  $\delta^{13}C_{DIC}$  for the full history of the industrial CO<sub>2</sub> release as well as the preceding preindustrial period back to the beginning of the Little Ice Age. This provides a wellfounded estimate of the anthropogenically uninfluenced, pre-industrial background level of surface water <sup>13</sup>C/<sup>12</sup>C ratios. Our records show small but systematic variations that appear to be linked to the climate fluctuations of the Little Ice Age.

Keywords: Coralline sponges; carbon isotopes; CO<sub>2</sub>; Caribbean; Little Ice Age; ocean surface water; industrial period; <sup>13</sup>C Suess effect.

# 1. Introduction

We investigate stable carbon isotope records of aragonitic skeletons of the coralline sponge *Ceratoporella nicholsoni* (Fig. 1), which lives in surface and upper thermocline waters of the Caribbean. These sponges precipitate aragonite very close to isotopic equilibrium with ambient sea-

water [*Druffel & Benavides*, 1986; *Böhm et al.*, 1996; *Reitner*, 1992]. Carbon isotope histories recorded by the sponges display the anthropogenic carbon isotope shift of the 19th and 20th century [*Druffel & Benavides*, 1986; *Böhm et al.*, 1996; *Swart et al.*, 1998; *Lazareth et al.*, 2000; *Moore et al.*, 2000; *Nozaki et al.* 1978] as well as variations prior to the 19th century. These preindustrial variations provide evidence for natural variations in the atmosphere/surface ocean  $CO_2$  inventory.

Based on ice core analyses [*Etheridge et al.*, 1996], it has been shown that the atmospheric  $CO_2$  mixing ratio (pCO<sub>2</sub>) varied by 6 ppm before the onset of industrial influences. Carbon isotope data of these ice core inclusions vary in concert with pCO<sub>2</sub> [*Francey et al.*, 1999]. These variations have been linked to climatic variations known as "Medieval Warm Period" and "Little Ice Age". Combined with the ice core record our data provides further constraints on the modeling of global climate changes during this period and supports the notion that the Little Ice Age had a measurable, global impact on the carbon cycle.

# 2. Material and Setting

We collected living sponges (Fig. 1) in 1993 and 1996 at Jamaica (Rio Bueno and Montego Bay) in reef caves in about 20 m of water and at the northwestern slope of Pedro Bank in 125 m of water (78°57'W 17°32'N). The sites are well exposed to the open sea. The Jamaican sites are situated within the mixed layer of the "Caribbean Surface Water", while the Pedro Bank site is positioned below a barrier layer [*Sprintall & Tomczak*, 1992] near the top of the "Subtropical Underwater" [*Morrison & Nowlin*, 1982]. Whereas the Caribbean Surface Water is mainly derived from the tropical South Atlantic, the Subtropical Underwater forms in the tropical and subtropical North Atlantic from where it is transported southwest into the Caribbean [*Schmitz & Richardson*, 1991]. Thus, the shallow water sponges formed their skeletons in waters that predominantly equilibrated with atmospheric CO<sub>2</sub> southeast of the Caribbean, while the deeper water sponges lived in waters that mainly reflect the atmosphere-surface water equilibration in the southwest flowing part of the North Atlantic subtropical gyre [*Schmitz & McCartney*, 1993].

As the deeper water site is positioned near the transition from Subtropical Underwater to Caribbean Surface Water, a varying degree of mixing between the two water masses can be expected at this water depth. Upwelling of Subtropical Underwater in the southern parts of the Caribbean [*Gordon*, 1967] leads to further mixing of the two water masses. It should also be noted that the surface water stratification pattern of the Caribbean may have been different during the Little Ice Age.

#### 3. Methods

Microsamples for isotope analysis were collected with a 0.5 mm dental drill and processed using standard procedures as described in *Böhm et al.* [1996]. Analyses were carried out at the isotope lab of the Geological Institute, University of Erlangen with a Finnigan Mat 252. Temporal resolutions of individual samples range from 2 to 4 years/sample, depending on the growth rates varying from 0.19 to 0.43 mm/year. External precision ( $2\sigma$ ) of  $\delta^{13}$ C mesurements based on multiple analyses (N=87) of standards NBS 19 and IAEA CO1 is ±0.04‰. All carbon isotope ratios are given in the common delta notation, relative to V-PDB (Vienna Pee Dee Belemnite).

Growth rates derive from U/Th dates in three specimens (measured at GEOMAR and at Göttingen University) and fits of <sup>14</sup>C AMS data (Fig. 2) to the radiocarbon "bomb spike" [*Benavides & Druffel*, 1986; *Druffel*, 1989] in specimen Ce145 [*Böhm et al.*, 1996] and specimen 392-19 (measured at the Leibniz-Labor, Kiel University). For the latter we assume that the timing

of the radiocarbon increase at 125 m depth was similar to the timing in surface waters of that region because the amplitude of change in  $\Delta^{14}$ C is similar to the surface water change (Fig. 2).

Based on these data we rely on steady growth rates to estimate the ages of individual sampled skeleton layers. Constant growth seems to be the rule, as growth rates calculated from U/Th dated sections of a single specimen show little variation, as long as homogeneous parts of a skeleton are considered (Fig. 1). We use linear regression to fit growth curves to the radiometrically dated tie points (Fig. 2). A linear growth model provides an optimal fit to the age data of specimen 392-19. We used a second order polynomial for specimen Ce95-2 and a third order polynomial for specimen Ce96-1. Details of the dating methods will be given elsewhere (Haase-Schramm, A., Böhm, F., Eisenhauer, A., Joachimksi, M.M., Dullo, W.-Chr., Hansen, B.T., Sr/Ca ratios and oxygen isotopes from sclerosponges: temperature and salinity history of the Caribbean mixed layer and thermocline during the Little Ice Age. Manuscript in preparation).

# 4. Results

All  $\delta^{13}$ C records (Appendix 1) show the full extent of the industrial decline (Fig. 3) caused by the anthropogenic addition of <sup>12</sup>C-enriched CO<sub>2</sub> to the atmosphere [*Druffel & Benavides*, 1986; *Keeling et al.*, 1979; *Friedli et al.*, 1986; *Francey et al.*, 1999; *Quay et al.*, 1992; *Laube-Lenfant et al.*, 1994; *Bacastow et al.*, 1996]. In all records the industrial period shows an older segment of slowly declining  $\delta^{13}$ C values. In contrast, the younger segment, starting about 1960 A.D., exhibits rapidly declining values (Fig. 3d-f). This parallels the records of atmospheric pCO<sub>2</sub> and its <sup>12</sup>C content, measured in ice core inclusions and air, which started to increase at higher rates between 1950 and 1960 A.D. [*Friedli et al.*, 1986; *Etheridge et al.*, 1996; *Francey et al.*, 1999] (Fig. 4).

The industrial decline in  $\delta^{13}$ C started in the first half of the 19th century after a short period of stable values around 1800 A.D. This late preindustrial plateau shows nearly constant values in all measured sponges (Fig. 3). The stable period was preceded by a slight decline in  $\delta^{13}$ C in the middle of the 18th century. The highest measured  $\delta^{13}$ C values of both shallow water records occur during the 17th and early 18th century (Fig. 3b,c). Before that period, i.e. before about 1600 A.D., values were in the same range as in the early 19th century. In the shallow water sponge records the mean  $\delta^{13}$ C of the 1400-1500 A.D. interval is significantly lower (p<0.01) than the 1620-1720 A.D. mean (Tab. 1).

Specimen Ce95-2 has two bio-eroded zones interrupting the isotope record (Fig. 3c). It shows a relatively high scatter in the pre-industrial part of the record. Two parallel isotope transects were sampled at a distance of about 1cm. We find an offset of about 0.1‰ between corresponding values of the two transects in the pre-industrial part (Fig. 3c). The second transect was sampled almost three years after the first one, therefore the observed  $\delta^{13}$ C offset may represent a similar ageing effect as previously described for sponge aragonite [*Druffel & Benavides*, 1986]. Nevertheless, the means of the 1400-1500 A.D. interval (transect 1: 4.92‰, transect 2: 4.81‰) and of the 1620-1720 A.D. interval (transect 1: 5.03‰, transect 2: 4.93‰) are again significantly different (p<<0.01) for both transects respectively. Furthermore, the means of the respective intervals from the first transect and sample Ce96-1 are identical (Tab. 1).

The deeper-water sample from Pedro Bank shows no obvious  $\delta^{13}$ C maximum during the 17th or 18th century, but rather very constant values in most of the pre-industrial interval (Fig. 3a). However, even in this sample we find a 1400-1500 A.D. mean that is slightly lower than the 1620-1720 A.D. mean (Tab. 1). The difference is significant at a 95% probability level.

# 5. Discussion 5.1. Preindustrial Variations

The sponge records provide the first preindustrial marine  $\delta^{13}$ C data with decadal resolution. Additionally, they cover the entire industrial period with higher accuracy, better temporal resolution and better timing constraints than previous records [*Druffel & Benavides*, 1986; *Böhm et al.*, 1996]. The excellent reproducibility between records from different sites of similar water depth (Fig. 3b, e) shows that sponge skeletons can reliably register regional environmental signals. The great similiarity between the Jamaica and Pedro Bank records indicates that the measured carbon isotope signal is representative for open sea surface waters of the central Caribbean and of the equatorial to tropical/subtropical Atlantic Ocean.

Surface water studies [*Bacastow et al.*, 1996; *McNichol & Druffel*, 1992; *Gruber et al.* 1998] show a high  $\delta^{13}C_{DIC}$  variability at time scales of days to months with amplitudes significantly greater than 0.1‰. Variations with periods of 3-5 years and amplitudes of about 0.1‰ have been observed on interannual time scales [*Bacastow et al.*, 1996]. Also,  $\Delta^{14}C$  in subtropical surface waters may vary significantly on subdecadal to decadal time scales while lacking the expected Suess effect long-term trend [*Druffel*, 1997]. Such subdecadal variations are explained by advection, vertical mixing and productivity changes. Longer surface water  $\delta^{13}C_{DIC}$  time series have not been available up to now. With the precision and resolution of our records (sample spacing about 2 to 6 years) we can reliably detect  $\delta^{13}C$  variations >0.05‰ on interdecadal and centennial time scales (>15-20 years). Our records (Fig. 3) demonstrate that variations in this time band are very small in the Caribbean. This is most obvious for the deeper-water setting at Pedro Bank, where the preindustrial (A.D. 1400-1850)  $\delta^{13}C$  variation is only ±0.07‰. Most of this variability is caused by a decrease of roughly 0.1‰ between A.D. 1730 and 1760 (Fig. 4).

The most prominent feature of the preindustrial shallow water record (Fig. 3b) is the 50 years long 0.1‰ increase in  $\delta^{13}$ C from about 1550 to 1600 A.D. and the subsequent decrease until about 1770 A.D. (Fig. 4).  $\delta^{13}$ C of atmospheric CO<sub>2</sub> increased by about 0.05 to 0.10‰ [*Francey et al.*, 1999] and atmospheric pCO<sub>2</sub> decreased by about 6 ppm [*Etheridge et al.*, 1996] during the same period (Fig. 4). Unfortunately, the coarse resolution and limited dating accuracy of both the sponge and ice core records currently prohibit conclusions about the precise temporal relation between the atmospheric and marine records.

The  $\delta^{13}$ C of atmospheric or marine dissolved total CO<sub>2</sub> is expected to correlate with 1/pCO<sub>2</sub> [*Keeling et al.*, 1979] as long as CO<sub>2</sub> is sequestered by or released from organic carbon. This is due to the strong <sup>13</sup>C depletion of organic carbon. Addition of such isotopically light carbon to the atmosphere/ocean inorganic carbon pool will in parallel increase the CO<sub>2</sub> and its <sup>12</sup>C content. There are, of course, other mechanisms to change atmospheric pCO<sub>2</sub> and the isotopic composition of CO<sub>2</sub> or DIC independently, e. g. changes in the ocean salinity or in the deepwater carbonate ion content will affect atmospheric pCO<sub>2</sub> without isotopic effects [*Broecker*, 1995].

The period of pCO<sub>2</sub> reduction and  $\delta^{13}$ C increase (1550-1700 A.D.) starts after the Spörer sunspot minimum and coincides with the Maunder sunspot minimum (approx. 1420-1540 and 1645-1715 A.D.; Fig. 4). These are periods of reduced solar activity [*Stuiver & Quay*, 1980; *Lean et al.*, 1995]. Moreover, the period between 1550 and 1850 A.D. was an interval of intermittent cool periods, called "Little Ice Age" [*Grove*, 1988; *Jones et al.*, 2001]. Thus, global cooling may have been the cause of the parallel pCO<sub>2</sub> reduction and atmospheric/marine  $\delta^{13}$ C increase [*Trudinger et al.*, 1999]. Increased solubility of CO<sub>2</sub> in colder surface waters or carbon sequestration in the terrestrial biosphere were discussed previously [*Etheridge et al.*, 1996, *Trudinger et al.*, 1999; *Joos et al.*, 1999]. Only the latter can explain the parallel atmospheric and marine  $\delta^{13}$ C increase.

Global cooling of sea surface temperatures would sequester atmospheric  $CO_2$  due to its higher solubility by about 10 ppm per kelvin. Models show that on time scales of some decades about half of this amount will be taken up by the oceans [*Bacastow*, 1996]. Thus, global sea surface

temperature cooling alone could explain the pCO<sub>2</sub> decrease, if the globally averaged cooling during the Little Ice Age was about 1K [*Grove*, 1988, *Trudinger et al.*, 1999, *Keigwin*, 1996, *Kitagawa & Matsumoto*, 1995; *Watanabe et al.*, 2001]. However, a global 1K cooling of sea surface temperatures would increase the isotopic equilibrium fractionation between air and water by about 0.1% [*Zhang et al.*, 1995] and this would only lower the  $\delta^{13}$ C of atmospheric CO<sub>2</sub> and leave the marine  $\delta^{13}C_{DIC}$  almost unchanged. This is because on a multidecadal timescale the exchangeable carbon mass in the ocean is much larger than the mass of carbon in the atmosphere [*Keir et al.*, 1998]. Therefore, the observed parallel increase of  $\delta^{13}C_{DIC}$  and of atmospheric  $\delta^{13}C$  during the period of lower pCO<sub>2</sub> requires an alternative explanation.

The observed close correlation of our data with atmospheric  $pCO_2$  indicates a common and therefore global cause. The most likely common cause of all observed carbon cycle changes is an increase in the global biogenic carbon mass during the Little Ice Age [*Trudinger et al.*, 1999; *Joos et al.*, 1999]. At the same time, the combined atmospheric and marine data allow for only minor global cooling of the ocean surface.

It is unknown, whether sequestration of organic carbon to the deep sea by increased oceanic productivity or rather increased storage of organic matter in terrestrial environments caused the observed changes in the earth's carbon budget during the Little Ice Age. Changes in the large scale ocean circulation [*Broecker*, 2000], but also increased wind induced upwelling in the southern Caribbean and subtropical North Atlantic [*Black et al.*, 1999; *deMenocal et al.*, 2000] may additionally have contributed to the observed variation of Caribbean  $\delta^{13}C_{DIC}$  during the Little Ice Age. Only additional long-term records of  $\delta^{13}C_{DIC}$  from other oceans will help to find out whether our observations represent a purely global signal. The only published Pacific Ocean carbon isotope record reaching back to the Little Ice Age of a coralline sponge from the Great Barrier Reef [*Wörheide et al.*, 1997] shows slightly higher values (about 0.05‰) during the 17th century, which is in accord with a global nature of the observed marine variations.

#### 5.2. Industrial Period

The sharp contrast between the almost invariant preindustrial record and the strong decline in  $\delta^{13}$ C since 1850 A.D. underlines the exceptional quality of this recent anthropogenic disturbance of the global carbon cycle. Comparing the observed decrease of  $\delta^{13}$ C in Caribbean surface water and in the atmosphere [*Francey et al.*, 1999], we find a consistent and significant decline in the ratio of the marine to the atmospheric change ( $\Delta \delta^{13}$ C<sub>DIC</sub>/ $\Delta \delta^{13}$ C<sub>Atm</sub>) from the pre-1960 period to the 1970s and 1980s (Tab. 1, Fig. 5): The marine  $\delta^{13}$ C decrease from preindustrial times to the 1960s was about 80% of the atmospheric change. The marine  $\delta^{13}$ C decrease from the 1960s to the 1980s, however, was only half the atmospheric change. This decline in the apparent isotopic equilibration between atmosphere and surface waters is evident in all four sponge records. Therefore, it is unlikely that this observation represents just an artefact of insufficient dating control and unnoticed decadal growth rate variations of our sponges.

In contrast to our observation of a reduced isotopic ocean-atmosphere equilibration after the 1960s, *Gruber et al.* [1999] report a  $\delta^{13}C_{DIC}$  decrease during the decade from 1983 to 1995 in the Sargasso Sea that equals or possibly exceeds the contemporary atmospheric  $\delta^{13}C$  decline. However, the youngest part of our isotope record from Specimen Ce95-2 points to a decrease of about 0.2±0.07‰ from the 1980s until 1995 (Fig. 3f). This decrease is compatible with the observations of *Gruber et al.* [1999]. Obviously, after the minimum of the 1970s (Fig. 5), the isotopic ocean-atmosphere equilibration increased again to the very high values of the 1990s reported by *Gruber et al.* [1999] and indicated by the youngest portion of our sponge records.

These variations of the isotopic ocean-atmosphere disequilibrium in the Caribbean Sea can be explained by the history of the atmospheric  $\delta^{13}C$ . The rate of decline of atmospheric  $\delta^{13}C$  increased

during the 1950s and 1960s, remained high until the late 1980s and was low during the 1990s [*Francey et al.*, 1999; *Gruber et al.*, 1999]. During the first half of the 20th century the carbon isotopic decline of the atmosphere was slow enough for the tropical surface ocean DIC to follow closely (about 80% "equilibration", i. e.  $\Delta \delta^{13}C_{DIC}/\Delta \delta^{13}C_{Atm}$ , in Caribbean and tropical North Atlantic surface waters, Tab. 1, Fig. 5). However, with the finite carbon isotopic equilibration time between atmosphere and surface waters, the faster atmospheric  $\delta^{13}C$  decrease during the 1960s and 1970s led to a lower degree of equilibration (about 50%). The subsequent slow down of the 1990s allowed the oceans to start catching up on the isotopic equilibrium (100-125% equilibration [*Gruber et al.*, 1999]; about 100% in Fig. 5). This varying degree of isotopic equilibration between atmospheric CO<sub>2</sub> and surface water DIC clearly reflects the current state of the global carbon cycle, far from steady state due to the serious anthropogenic interference of the last 200 years.

# 6. Conclusions

This study shows that the coralline sponge *Ceratoporella nicholsoni* is a very accurate recorder of  $\delta^{13}C_{DIC}$ . Coralline sponges are very useful in providing long-term, high-resolution  $\delta^{13}C$  records. These provide useful information for estimating ocean uptake of anthropogenic CO<sub>2</sub> [*Quay et al.*, 1992; *Bacastow et al.*, 1996; *Sonnerup et al.*, 1999, 2000] and the terrestrial biota turnover times [*Keir et al.*, 1998].

Our data shows a period of higher values during the Little Ice Age. These preindustrial variations are in concert with changes in the atmospheric carbon reservoir. Our data provides an independent evidence for variations in the global carbon reservoirs prior to the onset of anthropogenic perturbations.

Varying degrees of carbon isotopic equilibration between atmospheric  $CO_2$  and surface water DIC during the industrial increase of atmospheric p $CO_2$  reflect the dynamic response of the carbon cycle to human interference with magnitudes and rates of change that are unprecedented for the last 600 years and probably for the whole Holocene.

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# Figures

Fig.1: Polished slabs of the investigated *Ceratoporella nicholsoni* specimens (from top 392-19, Ce96-1 and Ce95-2) with sample transects (dotted lines). Uranium/Thorium ages are given as years of formation (A.D.) of the respective layers together with standard errors ( $2\sigma$ ). The 1965 datum of the top specimen is the radiocarbon bomb spike. The sampling spots of the U/Th datings and the radiocarbon sample that was assigned to the year 1965 A.D. are marked with bold black lines. Average growth rates are calculated from U/Th ages and the age of the surface, i .e the years of collection (1996 A.D. for Ce96-1 and 392-19 and 1995 A.D. for Ce95-2).

Fig. 2: a. Age depth curves of the three investigated sponge specimens. Depth is measured along the isotope sample transects. The data points at 0 mm represent the top surfaces of the sponges. Horizontal error bars represent the sample width (measured parallel to growth axis). BP= before present (i.e. before 1950 A.D.). The data points are fitted by polynomials.

b. Fit of the measured radiocarbon values of specimens 392-19 and Ce145 to the coral  $\Delta^{14}$ C curve for Atlantic Ocean surface waters east of southern Florida [*Druffel*, 1989]. Error bars are smaller than symbols except where shown. The width of the symbols is equivalent to the width of the samples. The lower x-axis corresponds to the Florida coral  $\Delta^{14}$ C curve. The upper x-axis shows the sample distance from the top of the sponge skeleton along the growth axis. We take the value intermediate between the pre-bomb and post-bomb levels to represent the year 1964±5 A.D. in both sponge skeletons. Spacing of the sponge radiocarbon samples is scaled to the lower x-axis (time) by assuming a growth rate of 0.19 mm/a. Specimen Ce145 probably grew slightly faster (0.22 mm/a [*Böhm et al.*, 1996]) than specimen 392-19.

Fig. 3: Stable carbon isotope records of the *Ceratoporella* skeletons. Error bars are  $\pm 2\sigma$  (standard deviation). The lines are locally weighted linear regressions (LOWESS, 10% window). Black arrows mark U/Th dates as shown in Fig. 1. Open arrows mark positions of the bomb spike in samples Ce145 [*Böhm et al.*, 1996] and 392-19. Hatched bars in panels c and f mark the positions of the bleached and bored zones in sample Ce95-2. Values from these bands may be slightly altered. Sample Ce145 is plotted with sample Ce96-1 for comparison.

Fig. 4: Comparison between Caribbean shallow (blue solid line) and deeper water (red dotted line) sponge  $\delta^{13}$ C records (inner left y-axis),  $\delta^{13}$ C of atmospheric CO<sub>2</sub> (green crosses, outer left y-axis) and atmospheric pCO<sub>2</sub> (circles, right y-axis, reciprocal scale). Atmosphere data are derived from Antarctic ice core and firn air inclusions [*Etheridge et al.*, 1996; *Francey et al.*, 1999] and air measurements (C. D. Keeling, Scripps: http://cdiac.esd.ornl.gov/trends/co2/sio-spl.htm). The atmospheric  $\delta^{13}$ C record is scaled for it's preindustrial mean and minimum values to fit the shallow water sponge record. The  $\delta^{13}$ C curves are smoothed by locally weighted linear regressions (9% window). The horizontal line marks the preindustrial (1400-1850 A.D.) means (280.5 ppm, -6.37‰, 4.95‰ VPDB respectively). Slight offsets in the timing between atmosphere and sponge records can be artefacts of the dating methods. Blue bar at the top shows the approximate duration of the Little Ice Age [*Grove*, 1988]. Red bars mark the Spörer (ca. 1420-1540 A.D.) and Maunder (1645-1715 A.D.) sunspot minima.

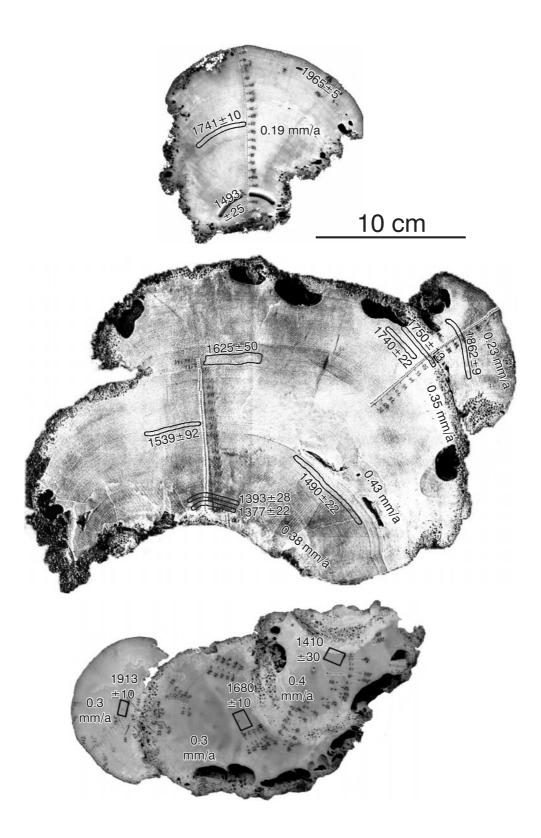
Fig. 5: Crossplot of carbon isotope values of Caribbean shallow water sponge aragonite and atmospheric  $CO_2$  [*Francey et al.*, 1999]. Age increases from left to right with increasing atmospheric values. The continuous line is a 4th-order polynomial fit to the data. The dashed line (right y-axis) is the first derivative of the polynomial fit. It represents the ratio of carbon isotopic change between Caribbean surface water DIC and atmospheric  $CO_2$ . This ratio varied around an average value of 0.8 before 1960, declined significantly during the 1960s and 1970s and reached

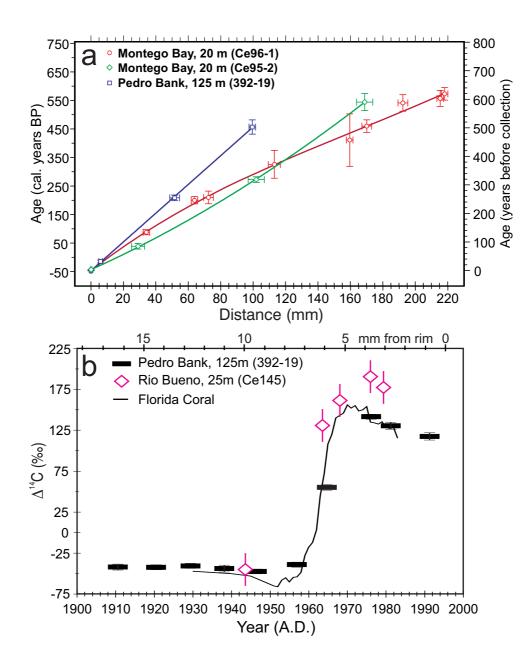
its highest values near 1 in the 1990s. a: Shallow water specimens from Jamaica. b: Deeper water specimen from Pedro Bank.

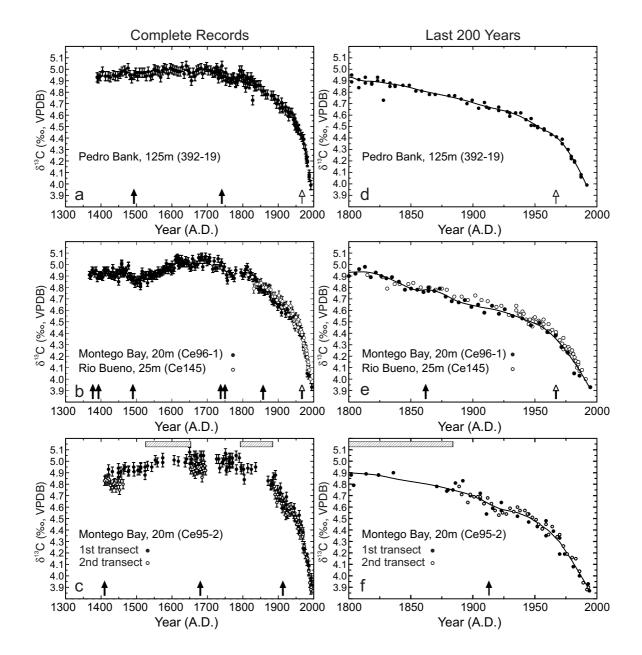
Tab. 1: Mean  $\delta^{13}$ C for certain time intervals calculated from the measured values. For sample Ce95-2 only data from the first transect are used. 1400-1850 means are calculated from the smoothed data sets interpolated to one year intervals. Decadal means are read from linear regression lines through respective decadal data. Preindustrial record of sample Ce95-2 is fragmentary, therefore the preindustrial mean may be slightly biased. Last four columns show measured marine decrease ( $\Delta\delta^{13}C_s$ ) from preindustrial mean (pi.) until 1960s and from 1960s to 1980s. Preindustrial mean for Ce145 is copied from Ce96-1. Last two columns give ratios between these values and the respective atmospheric carbon isotopic decline ( $\Delta\delta^{13}C_a$ , ice core and firn air inclusion data [*Francey et al.*, 1999]), which is pi. to 1960s: -0.67‰, 1960s to 1980s: -0.59‰.

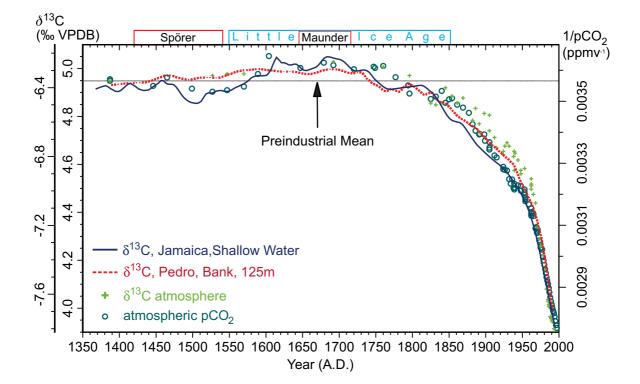
Appendix 1: Carbon isotope values (‰ VPDB) from Caribbean sponge specimens used in this study. Note that the ages given are only approximate estimates based on interpolations of U/Th dated layers!

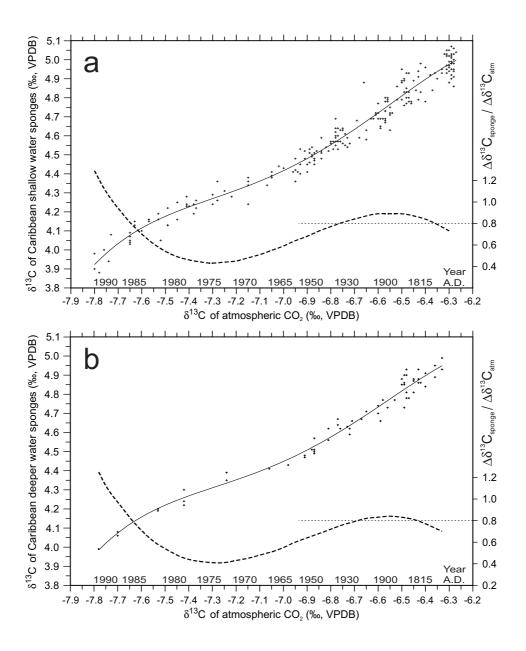
Sample	Mean $\delta^{13}$ C (‰ VPDB ±2 $\sigma$ / $\sqrt{n}$ ) of specified intervals (A.D.)						$\Delta \delta^{13} \mathbf{C_s}$		$\Delta \delta^{13} C_s / \Delta \delta^{13} C_a$	
Depth	1400-1500	1620-1720	1400-1850	1960-1970	1980-1990	pi60s	60s-80s	pi60s	60s-80s	
Ce96-1	4.92±0.01	5.02±0.01	4.95±0.01	4.37±0.03	4.06±0.13	-0.58	-0.31	0.9	0.5	
20 m	n=39	n=34	n=147	n=2	n=3	±0.03	±0.13	±0.09	±0.22	
Ce145 25 m			(4.95)	4.41±0.03 n=7	4.12±0.04 n=12	-0.54 ±0.03	-0.29 ±0.05	0.8 ±0.08	0.5 ±0.10	
Ce95-2	4.92±0.01	5.03±0.02	4.98±0.01	4.39±0.05	4.07±0.06	-0.59	-0.32	0.9	0.5	
20 m	n=13	n=16	n=71	n=7	n=7	±0.05	±0.08	±0.11	±0.13	
392-19	4.96±0.01	4.98±0.01	4.95±0.01	4.42±0.04	4.12±0.07	-0.53	-0.30	0.8	0.5	
125 m	n=17	n=20	n=107	n=4	n=4	±0.04	±0.08	±0.09	±0.14	











Ce145, 25 m		Ce96-1, 20 m				Ce95-2, 20 m, 1	st transect
	d13C	Year A.D. d13	c `	Year A.D. d			d13C
1986	4.07	1995	3.93	1612	5.05	1994	3.92
1985	4.13	1986	4.03	1610	5.04	1993	3.98
1985	4.15	1982	4.05	1606	4.97	1986	4.09
1984	4.10	1977	4.23	1604	4.95	1986	4.05
1983	4.16	1971	4.28	1601	5.00	1981	4.13
1982	4.19	1967	4.38	1598	4.94	1974	4.24
1981	4.22	1960	4.41	1595	4.98	1970	4.38
1980	4.25	1954	4.49	1593	4.98	1970	4.24
1979	4.24	1947	4.47	1590	4.95	1965	4.44
1978	4.22	1940	4.53	1587	4.97	1959	4.53
1977	4.23	1932	4.55	1585	5.01	1959	4.40
1977	4.28	1926	4.61	1582	4.95	1952	4.48
1976	4.24	1921	4.57	1580	4.96	1945	4.52
1974	4.29	1915	4.64	1577	4.96	1945	4.59
1973	4.26	1909	4.58	1575	4.94	1936	4.57
1972	4.31	1904	4.65	1572	4.95	1923	4.69
1970	4.34	1899	4.63	1570	4.93	1923	4.62
1967	4.41	1894	4.69	1568	4.93	1916	4.64
1964	4.43	1889	4.67	1565	4.92	1911	4.59
1962	4.41	1883	4.68	1561	4.95	1906	4.77
1961	4.42	1878	4.72	1557	4.90	1906	4.72
1957	4.48	1873	4.79	1554	4.92	1898	4.75
1956	4.45	1869	4.77	1552	4.90	1892	4.88
1954	4.50	1864	4.77	1550	4.91	1886	4.86
1953	4.51	1864	4.80	1547	4.91	1884	4.80
1951	4.50	1860	4.76	1544	4.88	1880	4.80
1949	4.53	1855	4.78	1542	4.92	1879	4.79
1948	4.46	1850	4.79	1539	4.90	1871	4.83
1946	4.52	1845	4.78	1537	4.93	1836	4.95
1945	4.51	1840	4.85	1534	4.91	1824	4.93
1943	4.52	1835	4.89	1531	4.84	1824	4.93
1941	4.57	1830	4.88	1528	4.91	1814	4.94
1938	4.56	1826	4.90	1525	4.90	1804	4.84
1937	4.57	1822	4.93	1522	4.88	1802	4.93
1935	4.64	1818	4.89	1519	4.91	1796	4.93
1991	4.00	1813	4.98	1519	4.89	1795	4.95
1988	4.08	1808	4.96	1516	4.94	1790	5.03
1983	4.16	1805	4.92	1514	4.89	1782	4.93
1982	4.16	1800	4.90	1511	4.83	1781	5.05
1978	4.26	1795	4.93	1511	4.81	1771	4.99
1973	4.36	1780	4.95	1509	4.87	1770	5.03
1967	4.39	1776	4.88	1505	4.87	1770	4.98
1961	4.48	1771	4.90	1503	4.84	1765	5.03
1957	4.52	1766	4.92	1499	4.86	1763	4.95
1950	4.54	1761	4.94	1496	4.83	1762	5.03
1945	4.48	1757	4.86	1494	4.89	1762	4.98
1940	4.59	1754	4.91	1491	4.86	1762	5.03
1935	4.60	1748	4.98	1488	4.89	1761	5.00
1929	4.56	1744	4.95	1485	4.87	1755	4.94
1923	4.64	1740	5.02	1483	4.90	1753	4.92

1919	4.63	1736	4.99	1480	4.90	1753	5.02
1912	4.68	1731	5.05	1478	4.99	1751	5.07
1907	4.69	1727	4.97	1478	4.99	1746	5.01
1902	4.73	1723	4.97	1475	4.89	1742	4.99
1896	4.69	1719	5.03	1472	4.95	1735	4.94
1891	4.72	1716	4.96	1469	4.93	1732	5.02
1885	4.72	1707	5.06	1466	4.94	1726	5.02
1879	4.78	1707	5.03	1463	4.92	1689	5.05
1875	4.79	1703	5.04	1460	4.95	1684	4.96
1870	4.78	1699	5.02	1460	4.93	1684	5.05
1865	4.80	1695	5.07	1458	5.00	1680	5.05
1858	4.80	1690	5.04	1458	5.01	1676	4.93
1852	4.84	1687	4.98	1455	4.92	1670	5.03
1848	4.83	1684	5.06	1452	4.95	1667	4.96
1841	4.83	1684	5.07	1450	4.85	1667	5.05
1837	4.83	1679	5.04	1447	4.92	1663	5.05
1831	4.83	1676	5.04	1447	4.92	1655	5.03
1827	4.79	1673	5.00	1443	4.93	1651	4.98
1815	4.91	1669	5.00	1442	4.89	1650	5.08
1015	4.91	1665	4.98	1440	4.89	1650	5.08
		1662	4.98 5.01	1430	4.90	1641	5.08
		1658	5.05	1433		1637	
		1656	5.05 4.98		4.90 4.91		4.99 5.00
				1428		1618	5.00
		1652	4.97	1425	4.91	1606	5.01
		1649	4.99	1422	4.91	1574	5.03
		1646	5.03	1420	4.95	1574	4.99
		1642	5.01	1416	4.95	1559	4.99
		1639	5.04	1413	4.93	1553	5.01
		1636	5.01	1411	4.94	1543	4.95
		1633	5.01	1408	4.95	1531	4.96
		1630	5.01	1405	4.89	1524	4.95
		1627	5.01	1402	4.90	1524	4.88
		1627	5.05	1399	4.91	1512	4.95
		1625	5.03	1397	4.90	1512	4.94
		1624	5.02	1393	4.91	1508	4.92
		1622	5.00	1390	4.88	1504	4.92
		1621	5.05	1386	4.95	1493	4.91
		1619	4.99	1383	4.92	1492	4.95
		1617	5.05	1378	4.94	1492	4.95
		1616	5.06	1375	4.95	1478	4.92
		1616	5.05	1371	4.90	1477	4.91
		1614	5.04	1368	4.91	1468	4.95
						1464	4.93
						1456	4.91
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3C 3.90 3.88 3.94 4.04 4.15 4.16 4.19 4.26	Year A.D. d13C 1992 1992 1987 1987 1982 1982 1982 1979	Yea 3.99 3.99 4.06 4.08 4.19 4.20	ar A. D. d13C 1758 1758 1752 1752 1752 1746	; 4.9 4.9 4.9 4.9
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4.26	1979	•	1746	4.9
		4.22	1741	5.0
	1979	4.24	1741	4.9
4.36	1977	4.30	1736	4.9
4.34	1972	4.35	1736	4.9
4.43	1972	4.39	1731	4.9
4.36				4.9
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				4.9
	1905			5.0
4.71	1899	4.73	1623	4.9
4.64	1895	4.70	1617	4.9
4.71	1891	4.74	1612	4.9
4.78	1885	4.77	1607	4.9
4.92	1881	4.77	1601	5.0
4.96	1870	4.78	1596	5.0
4.92	1865	4.78	1590	4.9
4.94	1859	4.81	1585	5.0
4.86	1854	4.81	1579	5.0
4.96	1849	4.86	1574	4.9
4.93	1849	4.86	1568	4.9
4.95				4.9
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				4.
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				4.9
	4.36 4.45 4.41 4.45 4.49 4.51 4.57 4.57 4.57 4.50 4.56 4.59 4.54 4.55 4.53 4.57 4.63 4.69 4.69 4.69 4.69 4.69 4.69 4.69 4.69	4.3619674.4519634.4119574.4519574.4919524.5119524.5119524.5419504.5719474.5719474.6019434.5519394.5919344.5419304.5519304.5519304.5319284.6319214.6319214.6319134.6919094.6919094.6919054.7118994.6418954.7118914.7818854.9218654.9418594.8618544.9618494.9518374.9218374.921837	4.36 $1967$ $4.41$ $4.45$ $1963$ $4.43$ $4.41$ $1957$ $4.48$ $4.45$ $1957$ $4.47$ $4.49$ $1952$ $4.50$ $4.51$ $1952$ $4.50$ $4.51$ $1952$ $4.49$ $4.54$ $1950$ $4.51$ $4.57$ $1947$ $4.57$ $4.57$ $1947$ $4.51$ $4.60$ $1943$ $4.56$ $4.56$ $1939$ $4.62$ $4.54$ $1930$ $4.62$ $4.55$ $1930$ $4.59$ $4.53$ $1928$ $4.63$ $4.55$ $1930$ $4.59$ $4.53$ $1921$ $4.67$ $4.68$ $1921$ $4.64$ $4.63$ $1913$ $4.66$ $4.69$ $1911$ $4.67$ $4.69$ $1909$ $4.71$ $4.69$ $1905$ $4.66$ $4.71$ $1899$ $4.73$ $4.64$ $1895$ $4.70$ $4.71$ $1891$ $4.74$ $4.78$ $1885$ $4.77$ $4.92$ $1865$ $4.78$ $4.94$ $1859$ $4.81$ $4.96$ $1849$ $4.86$ $4.93$ $1849$ $4.86$ $4.94$ $1844$ $4.86$ $4.95$ $1837$ $4.85$ $4.92$ $1837$ $4.85$	4.361967 $4.41$ 1731 $4.45$ 1963 $4.43$ 1727 $4.44$ 1957 $4.48$ 1727 $4.45$ 1957 $4.47$ 1721 $4.45$ 1957 $4.47$ 1721 $4.45$ 1952 $4.50$ 1721 $4.51$ 1952 $4.49$ 1715 $4.54$ 1950 $4.51$ 1709 $4.57$ 1947 $4.57$ 1703 $4.57$ 1947 $4.51$ 1698 $4.60$ 1943 $4.56$ 1693 $4.56$ 1939 $4.62$ 1687 $4.59$ 1934 $4.62$ 1682 $4.54$ 1930 $4.62$ 1675 $4.55$ 1930 $4.59$ 1670 $4.53$ 1928 $4.63$ 1660 $4.63$ 1921 $4.64$ 1650 $4.63$ 1921 $4.64$ 1650 $4.64$ 1939 $4.71$ 1635 $4.69$ 1911 $4.67$ 1640 $4.69$ 1905 $4.66$ 1629 $4.71$ 1899 $4.73$ 1623 $4.64$ 1855 $4.77$ 1607 $4.71$ 1891 $4.74$ 1612 $4.78$ 1885 $4.77$ 1601 $4.92$ 1844 $4.86$ 1559 $4.94$ 1849 $4.86$ 1568 $4.95$ 1844 $4.86$ 1568 $4.94$ 1844 $4.86$ 1559 $4.95$ 1837 $4.85$ 1550 $4.92$ 1833 $4.88$ 1545

1664	4.94	1828	4.90	1533	4.98
1661	4.91	1828	4.73	1528	4.94
1659	4.93	1823	4.93	1522	4.97
1657	4.95	1823	4.90	1516	4.97
1655	4.96	1818	4.87	1509	4.97
1653	4.93	1818	4.88	1504	4.94
1462	4.80	1813	4.93	1498	4.97
1455	4.85	1813	4.88	1493	4.95
1452	4.75	1808	4.91	1487	4.92
1445	4.80	1808	4.84	1482	4.9
1442	4.77	1802	4.95	1477	5.0
1439	4.81	1802	4.89	1471	4.9
1435	4.80	1797	4.99	1467	4.9
1432	4.77	1797	4.93	1460	4.9
1427	4.79	1791	4.94	1454	4.9
1423	4.84	1791	4.99	1448	4.9
1420	4.85	1785	4.92	1441	4.9
1417	4.83	1785	4.90	1435	4.9
1413	4.84	1780	4.89	1426	4.9
1410	4.83	1780	4.88	1421	4.9
		1774	4.93	1413	4.9
		1774	4.92	1404	4.9
		1769	4.92	1398	4.9
		1769	4.91	1392	4.9
		1763	4.89	1389	4.9
		1763	4.89		