

**A global view of coral reef cementation as a function of seawater
aragonite saturation states**

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ABSTRACT

Throughout the past decade the findings of many research studies have led to ample concern about rising atmospheric CO₂. As atmospheric CO₂ increases a process called ocean acidification begins to take place. Ocean acidification causes major changes in the carbonate chemistry of the ocean as well as the vital biological and geochemical processes of the sea. Oceanic uptake of CO₂ drives seawater pH to become more acidic as well as decreasing the saturation state of calcite and aragonite, both of which are critical minerals in the coral reef ecosystem. CaCO₃ minerals can either be produced by calcifying reefal organisms such as coral, or can be inorganically precipitated out of seawater as cements. Both processes help to bind reef framework and protect reefs against erosion. Ocean acidification is thought to decrease calcification rates in coral reefs and increase susceptibility to erosion. Numerous researchers have studied the effects of low pH on calcifying organisms as well as the effects of changing nutrient and saturation state levels on calcifying organisms. What has been over looked is the affect of ocean acidification on the relative abundance of carbonate cements that help bind reefs. This study used samples of corals collected from reefs around the world and quantified the amount of intra-skeletal pores in those corals that contained cements as a proxy for reef cementation. An empirical relationship between decreasing inorganic CaCO₃ cement abundances and decreasing aragonite saturation, predicts decreasing cementation in a high CO₂ world. Salinity serves as a promising predictor of inorganic cementation as well.

INTRODUCTION

Overview

At present historically high amounts of carbon dioxide are being released into the atmosphere (IPCC 2007). Combustion of fossil fuels, as well as interactions between humans and the environment are significant contributors to this trend (IPCC 2007). The ocean absorbs up to one third of the carbon dioxide present in the Earth's atmosphere (Kleypas et al. 2006), and when atmospheric CO₂ is increasing a process called ocean acidification begins to take place. The effect of contemporary ocean acidification on the ocean's chemistry, as well as the ocean's biological systems, is an area of ongoing research.

Ocean acidification is a change in ocean chemistry due to the absorption of atmospheric carbon, phosphate, and sulfur compounds (Doney et al. 2007). Through carbon speciation and the reactions between gaseous carbon dioxide and water, ocean pH becomes increasingly acidic as these gases are added to seawater. Increasing carbon dioxide is the major problem. The initial chemical reaction involving CO₂ and H₂O produces carbonic acid (H₂CO₃). Carbonic acid then dissociates into hydrogen ions and bicarbonate (Doney et al. 2009). The dissociation of bicarbonate produces an even higher concentration of hydrogen ions and as well as carbonate ions (Doney et al. 2009). The large concentration of hydrogen ions lowers the pH of the surrounding ocean water. Simultaneously, the conversion of gaseous CO₂ into bicarbonate and carbonate effectively reduces the dissolved CO₂ concentration in the ocean, therefore allowing more diffusion from the atmosphere (Doney et al. 2009). This feedback causes ocean pH levels to continually decrease (Fig. 1).

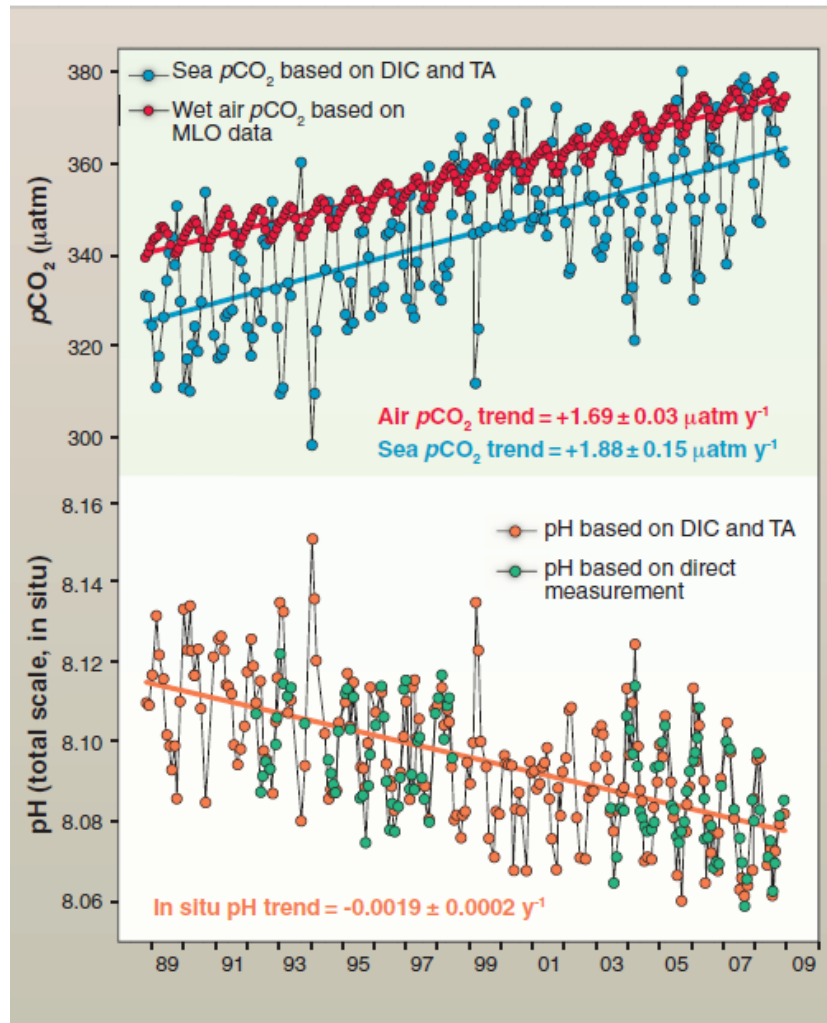


Fig. 1. Time progression of $p\text{CO}_2$ (top) and pH (bottom) of seawater observed over a period of 20 years at Mauna Loa Observatory, Hawaii. As seawater $p\text{CO}_2$ increases pH becomes more acidic (Doney et al. 2010).

One of the most significant repercussions of ocean acidification is that it is expected to reduce coral reef calcification and increase reef susceptibility to erosion (Manzello 2008). As ocean pH becomes acidic the saturation state of the mineral aragonite decreases (Fig. 2, Doney et al. 2008). This occurs because as seawater's pH decreases (becomes more acidic) the carbonate ion concentration in the oceans decreases. Aragonite is the mineral produced by most calcifying reefal organisms, in particularly corals. The saturation state is a measure of the thermodynamic

drive for a mineral to precipitate or dissolve (Doney et al. 2008). As the aragonite saturation state lowers, reef forming organisms should either produce less CaCO_3 per cm^3 shell (less dense shells) or less shell entirely (Ries et al. 2009). The CaCO_3 shells of reef organisms provide protection, buoyancy regulation, anchoring to substrates, and light modification (Kleypas et al. 2006). Thus, less calcification due to ocean acidification should have negative effects on calcifying organisms. To date, the greatest effect from decreasing calcification rates is less dense coral structures which, as a result, promotes bioerosion and reduces stability of reef framework (Manzello et al. 2008).

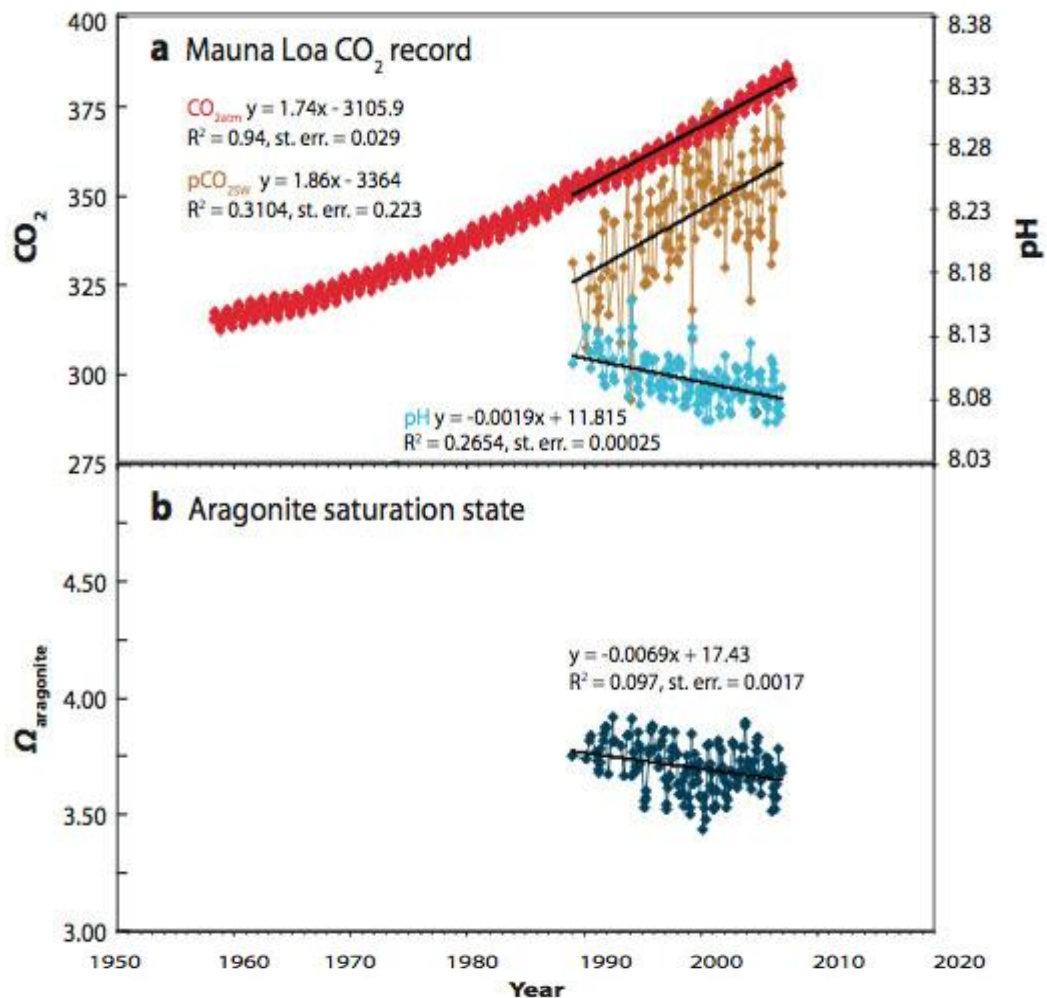


Fig. 2. Time series of atmospheric pCO₂ (red) and sea water pCO₂ (yellow), sea surface pH (light blue), and aragonite saturation state (dark blue). There is a decrease in both sea surface pH and aragonite saturation states with increasing pCO₂ over time (Doney et al. 2008).

It is important to distinguish between biogenic CaCO₃ (produced by calcifying organisms) and inorganic CaCO₃ cements (precipitated out of seawater). CaCO₃ produced by organisms is vital in creating reef stability and expansion, but the precipitation of secondary CaCO₃ from seawater also serves to bind reef framework and protect against erosion (Perry and Hepburn 2008). Ocean acidification should affect both forms of subsea CaCO₃ production.

Various research studies have been focused on ocean acidification because of the large geochemical and biological effects that result from acidification. At present researchers have studied calcifying organism's response to a decrease in ocean pH (e.g. Andersson and Kuffner 2009; Ries et al. 2009) as well as the individual effects of aragonite saturation state, nutrient influx, and temperature on calcifying organisms (e.g., Silverman et al. 2007). What has been overlooked is the role of inorganic cementation. Aside from Manzello et al. (2008), no one has considered the effect of acidification on the relative abundance of carbonate cements that help bind reefs (Rasser 2002). A key question that encompasses ocean acidification research is the uncertainty surrounding the production of cements in a high CO₂ world. Will there be a decrease in cement production, and if so, how much less will be produced?

Coral reefs

All reef systems can be broken up in to subzones, each of which has distinctive physiological and ecological characteristics that relate to the amount of cementation that is produced in that zone (James 1990). The primary zones that comprise most coral reef systems are the back reef, reef crest, and fore-reef (Fig. 3). The growth of each of these zones varies substantially between geographic areas. In certain locations there may be a gradual transition

between zones while in other localities there is a sharp, sudden transition (NOAA Coral Reef Conservation Program 2010). The characteristics of each zone depend on the water level, light penetration, water movement, sedimentation and numerous other factors that can affect both the biology and geochemistry of the water (NOAA Coral Reef Conservation Program 2010). These factors also contribute to producing two types of sediment accumulations.

Framework accumulations in the reef crest and fore-reef are dominated by branching coral and coral heads (Marshall 1985) and are also characterized by subsea cementation that helps bind reef framework and provide the reef with resistance to erosion (Manzello 2008). As one progresses landward from the reef crest the presence of framework accumulations becomes less abundant and the occurrence of unconsolidated material increases (Marshall 1985). Detrital accumulations are broken and physically reworked, sand to grain sized sediment and shell fragments (Marshall 1985) and contain no *insitu* coral framework.

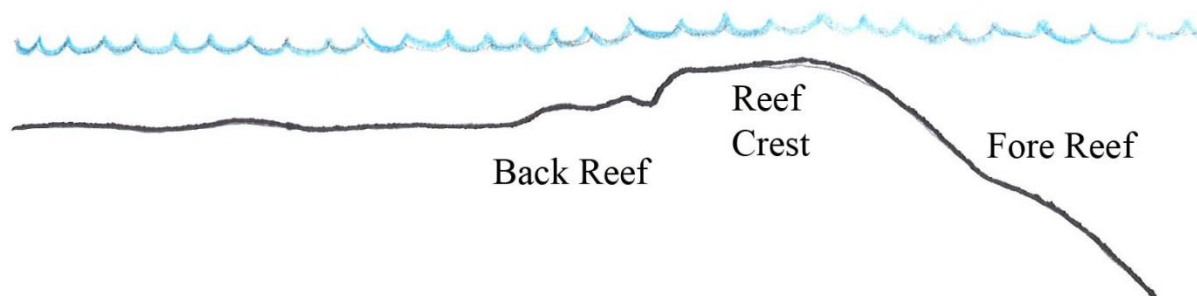


Fig. 3. Diagram showing coral reef zones.

The back reef consists of shallow waters that occur closest to the shoreline and experience the largest variations in temperature and salinity (NOAA Coral Reef Conservation Program 2010). Shallow waters and the intermittent exposure of the back reef during low tides limit the growth and expansion of coral (NOAA Coral Reef Conservation Program 2010). Because of the limited coral growth, detrital accumulations dominate (Perry and Hepburn 2008). Reef deposits in this section generally have little to no carbonate inter-skeletal or intra-skeletal cementation (Marshall 1985). Rarely there are secluded bands of acicular aragonite and peloidal micrite (Perry and Hepburn 2007).

The reef crest is where a solid, well-lithified encrusted framework accumulation develops (Marshall 1985). Encrustors aid in forming that reef framework by developing thick layers of calcareous crusts on coral substrates (Riegal 2002). Encrustors include conical and globose forms of foraminifera, bryozoans, serpulids, red algae, and sclerosponges (Perry and Hepburn 2007). Other than the biogenic CaCO_3 produced by encrustors, there is also abundant amounts of cementation in the reef crest. Typically there are peloidal micritic cements around inter-skeletal clasts as well as botryoids of aragonite cements that develop between encruster sheets (Perry and Hepburn 2007). Acicular aragonite needles can be seen in both intra- and inter-skeletal cavities (Behairy and El-Sayed 1984). The extent to which cementation occurs on the reef crest has been documented by Marshall (1985) and Lighty (1985). Samples examined at the outer shelf, including the reef crest by Marshall, had “every available pore or boring completely infilled,” including both inter and intra skeletal voids. The reef crest is considered the most productive subzone of the reef as far as CaCO_3 production and cementation (James and Choquette 1984).

The second most productive subzone of the reef is the fore-reef. Surface encrustation in this zone includes bryozoans, serpulids, and coralline algae (Perry and Hepburn 2008). Sponges also can coat large areas of coral substrata (Perry and Hepburn 2008). One key feature regarding this region of the reef is the continual bioerosion and cementation process which allows for the quick infilling of reef framework (Riegal 2002). Vacated borings are cemented by peloidal micrite as well as botryoidal aragonite cements (Perry and Hepburn 2008). Both micrite and aragonite can also occur in vacant framework cavities as well as in unoccupied pores within clasts (Perry and Hepburn 2008).

Global distribution of corals and reef cements

Inorganic and biogenic calcium carbonate is most abundant in shallow water environments in the tropics and sub-tropics. The narrow and consistent environmental conditions required for coral growth and expansion limit reefs to no higher 30° latitude (NOAA Coral Reef Conservation Program 2010). Large amounts of marine cements and non-skeletal grains such as ooids and peloids are predominately restricted to the tropics (James 1990 and Milliman 1974). The one exception is Bermuda, located at 32°N latitude, because it is located within warm Gulf Stream waters (Barnes 1987). Generally, the diversity of reef assemblages' declines with increasing latitude and most carbonate production at mid to high latitudes is high-Mg calcite or calcite with little to no aragonite (James 1990). Latitudinal differences in the geochemistry of the seawater are a factor contributing to this trend. Aragonite saturation states decrease with increasing latitude (Fig. 4) due to temperatures effect on $p\text{CO}_2$. With decreasing seawater temperature the solubility of CO_2 in seawater increases. The latitudinal zonation of aragonite saturation also explains why subsea cements are generally restricted to the tropics. Regions such as the Bahamas and the Red Sea are distinguished by some of the world's highest saturation

states (>4.0), whereas the eastern tropical Pacific, primarily the Galapagos, is typified by some of the most depleted saturation states in the world (<3.0).

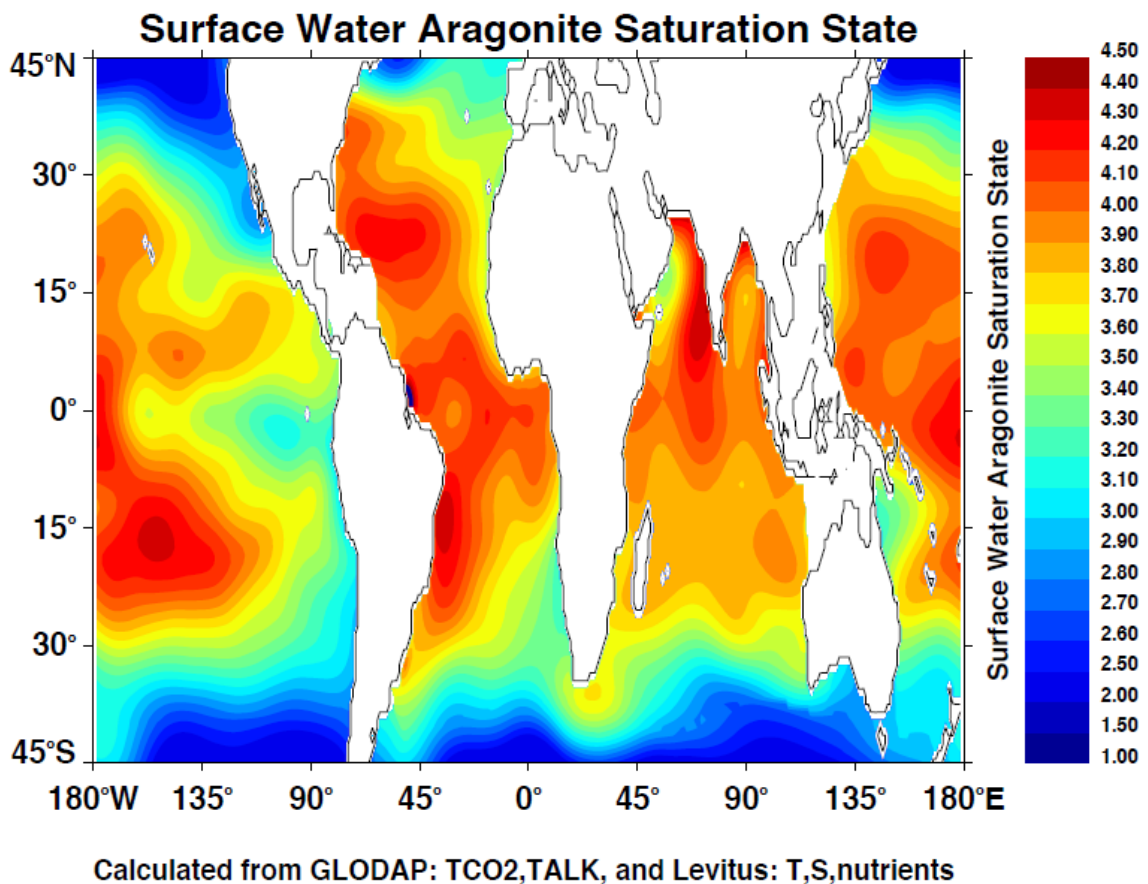


Fig. 4. Distribution of aragonite saturation states (Kleypas 2010).

The Research Problem

Understanding the fluctuation of subsea cementation relative to geographic location and spatial variation within the reef setting provides the necessary context for the study at hand. The goal is to perform a global survey of marine cementation within coral skeletal porosity as a function of seawater aragonite saturation states, and assess whether an empirical relationship

exists between the extent of cementation and saturation state, alkalinity, and $p\text{CO}_2$. Such an empirical relationship could then be applied in the creation of a model to predict future reef cementation in a high CO_2 world where ocean pH and aragonite saturation levels are at historic lows.

METHODS AND MATERIALS

Sample Provenance

Sampling and quantifying cementation in coral reefs is not an easy task given the large variety of void spaces that exist in reefs (e.g, within individual skeletons, between skeletal components, and in large constructional voids). To simplify the issue, it was assumed that cementation in the intra-skeletal pores of individual corals was a practical proxy for the amount of cementation in the reef as a whole. Such an assumption was used successfully by Manzello et al. (2008). Accordingly, efforts were made to collect coral samples from reefs across the entire globe.

Coral samples from the Caribbean, the eastern tropical Pacific, the Ryukyu Islands of Japan, the Great Barrier Reef, four sites across the western equatorial Pacific, and two sites in the Indian Ocean were utilized for this study. No field work was done in the context of this study; all samples were provided by Dr. Joanie Kleypas (NOAA), Dr. Derek Manzello (NOAA), Dr. Julie Cole (University of Arizona), and Dr. Yasu Iryu (Nagoya University, Japan). These sites span the current range of ocean pH and saturation states (Fig. 4 and 5) with the eastern tropical Pacific being a location of currently low pH (acidified ocean analogs), and the Caribbean being a location with current high pH (pre-acidification analogs). Samples differed among sites, ranging from coral pieces to framework debris, or in the case of Dr. Cole's corals cores were taken

through large Porites heads. All examined material was estimated or known by its collectors to have been ~25-40 years old prior to collection.

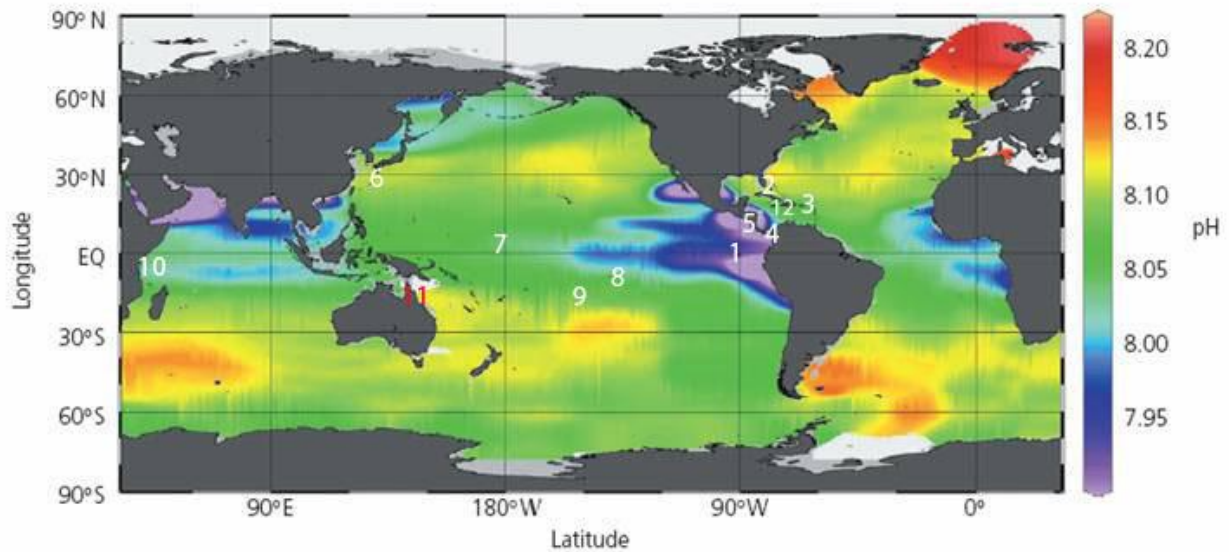


Fig. 5. Global distribution of coral reef samples and the pH representative at that location (Lenton et al. 2009). Numbers refer to sample sites (see Table 1).

Site number	Location
1	Galápagos
2	Key Largo
3	Puerto Rico
4	Gulf of Panamá
5	Gulf of Chiriquí
6	Ryukyu Islands
7	Kiribati
8	Marquesas
9	Tahiti
10	Tanzania
11	Great Barrier Reef
12	Bahamas

Table 1. Site number and corresponding location from Fig. 5

Representing both high and low pH waters, coral samples from Key Largo, Puerto Rico, the Galapagos Islands, and Panama were provided by Dr. Derek Manzello (NOAA). Both the Galapagos and Panama samples were used in prior studies (Manzello et al. 2008 and Manzello 2010). The samples collected in Panama, Puerto Rico, and Key Largo are part of *insitu* reef framework. The Galapagos samples are a blend of reef cobbles and reef framework and come from the lowest pH waters of all the samples collected. The rapid bioerosion of framework structures in the Galapagos (Manzello 2008) only allowed for one location of *insitu* reef framework to be sampled (Devil's Crown, Floreana Island). Key Largo samples are separated into inner shelf and outer self with ~5km between collection sites (Fig. 6).

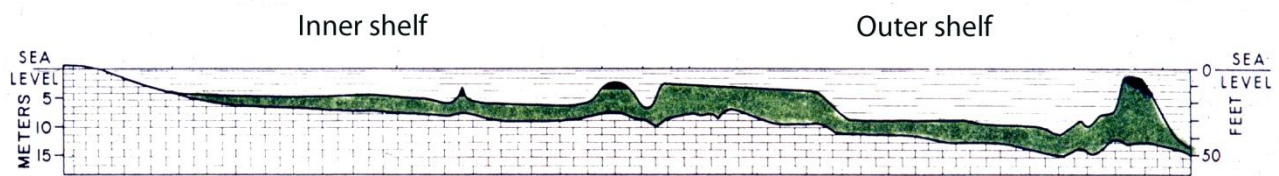


Fig. 6. Florida profile showing the distance between the inner and outer shelf. The figure spans 7.3km (Enos and Perkins 1977). Black denotes reef.

For regions of intermediate aragonite saturation states, samples collected in the Ryukyu Islands of Japan were provided by Dr. Yasu Iryu (Nagoya University, Japan). None of these samples had been studied by prior workers, and they were collected between August 1998 and November 2007. All samples are reef cobbles collected on the reef and reef flat and are not considered reef framework.

All other samples were derived from high pH oceanic settings. These include Tanzania, Kiribati, Marquesas, Tahiti, and the Great Barrier Reef samples provided by Dr. Julie Cole (University of Arizona). All samples were obtained from large *Porites* colonies. These samples

were acquired with a submersible drill that cored from the top of a coral head downward through the seasonal growth bands. The seasonal growth bands permit ages through the coral heads to be determined. Samples for this study were collected from 25-40 year old bands. All samples provided by Dr. Cole are considered reef framework.

Following the collection of coral from around the globe, seawater geochemical data was required for the relationship between aragonite saturation states and cement abundance in corals to be tested. For each specific site alkalinity and aragonite saturation data was collected from the on-line GLODAP(Global Ocean Data Analysis Project) data base, the World Ocean Atlas or from published research papers specific to each area of study (e.g., Manzello 2010). The one exception was the sites of Key Largo, FL and Puerto Rico where Derek Manzello provided unpublished onsite geochemical data.

There was significant CO₂ system variability across all sites, especially in terms of seasonality, because of metrological and oceanographic changes throughout the year. For instance, the aragonite saturation state for the Galapagos saw a variation of 0.61 between dry and wet seasons (higher in the wet and lower in the dry). Because of this variability, the use of annual averages of geochemical data for a location was applied. Cementation occurs over an interval of time where as geochemical data is sampled at one point in time, thus using an annual average is more suitable in terms of the time interval required for cementation.

Quantifying cement

Quantification of the amount of cement was done by the method of Manzello et al. (2008). Thin sections were made from each coral sample taken from the different localities across the globe. Four to five evenly spaced transects across the thin section were defined. Along

each transect the total number of pores with and without cement was counted. The total amount of cement bearing pores is then expressed as a percentage.

A significant step to quantifying the amount of cement was being able to identify the petrographic characteristics of cement in thin section. Shallow marine cement in reefs and corals typically consists of aragonite (orthorhombic CaCO_3). Aragonite precipitates are generally seen as needles a few micrometers to tens of micrometers in length (James 1990). The needles can occur in an array of different habits (Fig. 7) including columnar, acicular and prismatic. According to James (1990) the primary habits seen in reef material are (1) isopachous rinds, occurring as epitaxial growths on substrates such as corals. (2) An intergranular mesh of crystals focused around a grain surface in a random orientation so that any vacant space around the grain is consumed. (3) Botryoidal habit which is a globular or fan shaped arrangement of cement around a grain or group of grains. Acicular and botryoidal habits are most commonly seen and generally occur in cylindrical bivalve borings, macroboring from sponges, and in vacant intra-skeletal pores of corals (Ginsburg and James 1976). Aragonite can occur as both intergranular and intragranular cement.

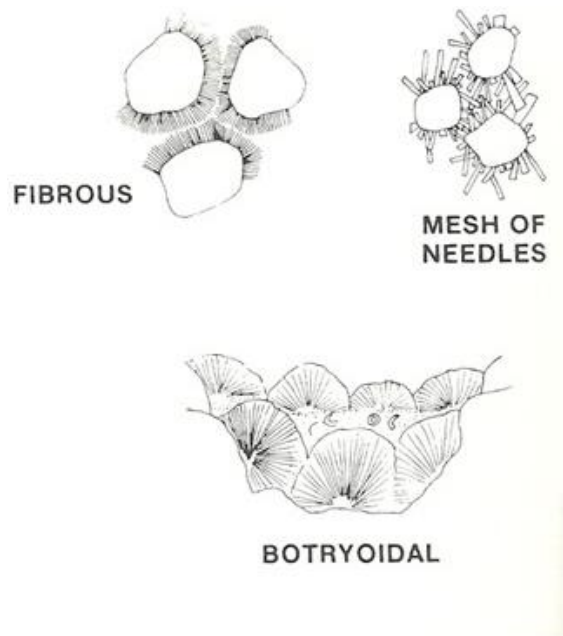


Fig. 7. The primary habits of aragonite cements in modern carbonates (James 1990).

Crusts of micritic, sparry, and peloidal high-Mg calcite cements (Fig 8) can also occur in corals and reefs (James 1990), but none of the cement habits were noted in any sample studied herein.

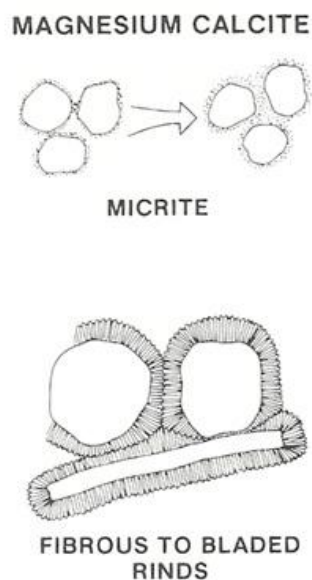


Fig. 8. Common habits of magnesium calcite cements in modern carbonates (James 1990).

RESULTS

Common cavity infillings

Although botryoids and acicular aragonite were some of the more eye catching infillings observed, they were not the only inorganic aragonite cement present in intra-skeletal pores. Numerous samples also contained red algae, spicules, and sediment (Fig. 9). In most samples algae was seen encrusting the edges of corals and outermost pores, while spicules and sediment were spatially variable throughout pores in the entire sample. It was not uncommon for sediment and cements to be present in the same intra-skeletal pore and in all cases the distribution of cements in coral porosity was extremely variable.

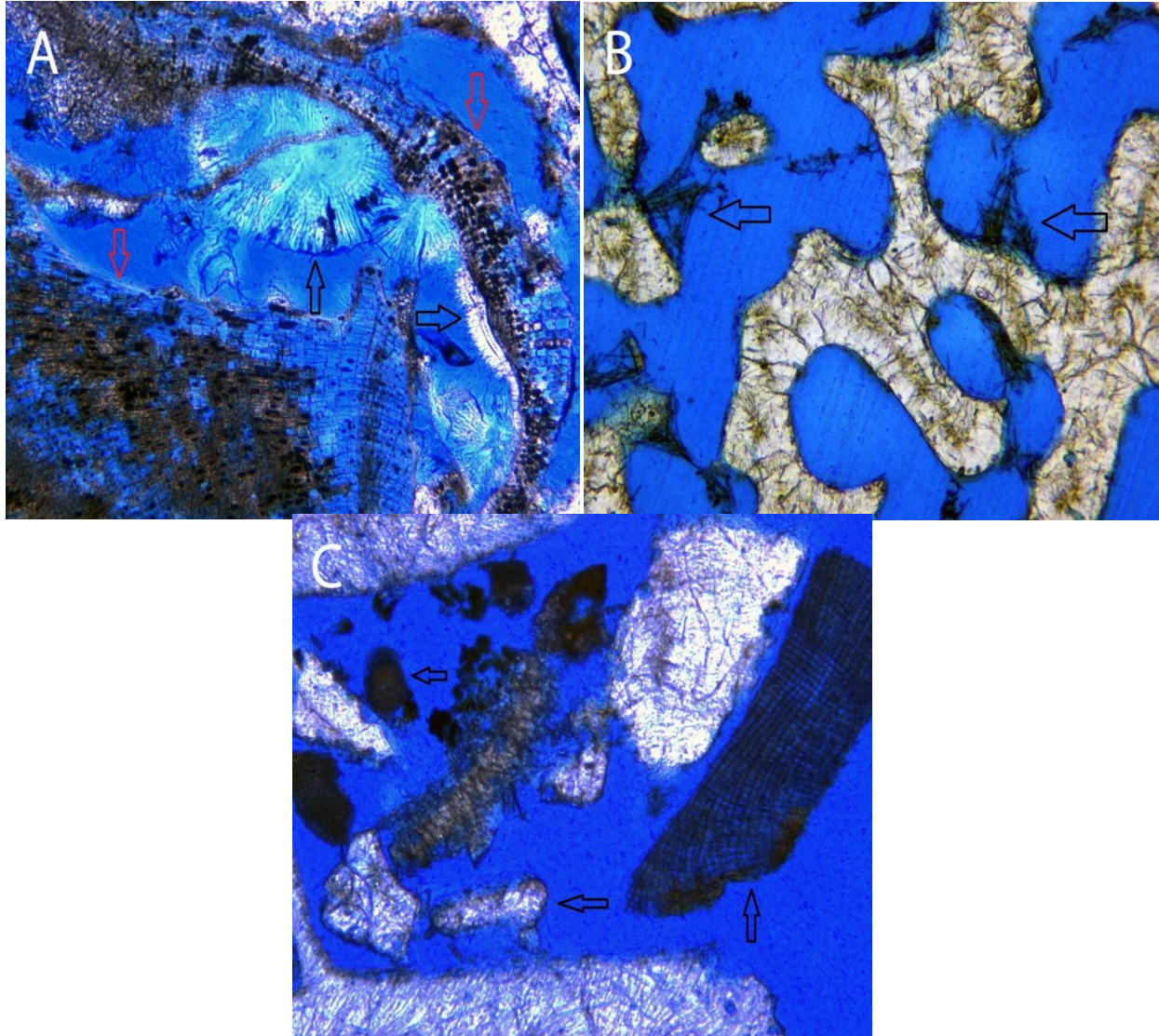


Fig. 9. Thin section photomicrographs of additional material within intra-skeletal pores. (A) Sample from the Gulf of Chiriquí, Panama with encrusting red algae (red arrows) along with botryoidal and acicular aragonite (black arrows). (B) Sample from Santa Fe Island, Galápagos with masses of spicules (a skeletal element of most sponges). (C) An example of micritic sediment, red alga, and detrital coral fragments in the same skeletal void from Santiago Island, Galápagos.

Cement abundance

All data on cement abundance is presented in Table 2. As found in Manzello et al. (2008), locations in Panamá had minimal amounts of cementation. On average, only 9.7% and 3.5% of intraskeletal pores contain cement in the Gulf of Chiriquí and Gulf of Panamá corals, respectively. Nearly all intra-skeletal pores were lacking cement and in no case involving both the Gulf of Chiriquí and the Gulf of Panamá did cementation completely fill an intra-skeletal cavity. The greater amount of cement in Panamá occurs in the Gulf of Chiriquí which does have a higher average aragonite saturation state than the Gulf of Panamá sites.

The amount of pores containing cement in Galápagos samples averages 7.3% which is slightly higher than the average of 4.4% reported by Manzello et al. (2008). As this study shared many Galapagos samples with the Manzello et al. study, this result was surprising. However, it occurs primarily because this study included more framework samples from Devil's Crown, which contained more cement (on average 11.6% of pores have cement).

The highest cement abundance was seen in coral reef framework collected on the outer shelf of Key Largo, Florida. Approximately 20% of intra-skeletal pores were seen to have cement present. Key Largo inner shelf corals had an average of 13.0%. Although it was expected that Key Largo outer shelf was to have more cement in comparison to the inner shelf, the aragonite saturation state for the inner shelf was 14% higher than the outer shelf. This result was surprising because the outer shelf is expected have higher saturation states than the inner shelf because the outer shelf is exposed to open ocean alkalinities which would naturally raise the saturation state.

Location	Reef framework	Location in reef	pH	Salinity	TA ($\mu\text{eq Kg}^{-1}$)	TCO ₂ ($\mu\text{mol Kg}^{-1}$)	pCO ₂ (Pa)	Arag. saturation state	Average Percent cement	N	Reference
Key Largo-inner	Framework		8.17	36.5	2327	1925	27.2	4.40	13.0	10	Manzello, personal communication
Key Largo-outer	Framework		8.07	36.2	2386	2045	37.8	3.80	19.7	10	Manzello, personal communication
Puerto Rico	Framework	fore-reef		36.2				3.60	15.1	13	Manzello, personal communication
Galapagos Is. Devil's Crown	Framework		7.98	34.9	2316	2073	47.9	3.06	11.6	6	Manzello et al. 2010
Galapagos Is. (excluding Devil's Crown)	NOT		7.91	35.0	2309	2073	58.5	2.74	8.14	11	Manzello et al. 2010
Bahamas	Framework		8.07	36.4	2382	2028	37.2	4.00	13.0	1	Manzello et al. 2008
Panamá - G. of Chiriquí	Framework	fore reef	8.04	32.0	2093	1797	41.0	3.53	9.70	18	Manzello et al. 2010
Panamá - G. of Panamá	Framework	reef flat /fore reef	8.04	29.7	1969	1717	36.7	2.98	3.52	6	Manzello et al. 2008
Ryukyu Islands, Japan	NOT	reef crest/fore reef	8.11	34.6	2295	1960		3.95	10.9	12	Kleypas, GLODAP, World Ocean Atlas 2001, Feely (2009)
Great Barrier Reef	Framework		8.13	35.3	2330	1975		3.60	5.54	1	Kleypas, GLODAP, World Ocean Atlas 2001, Feely (2009)
Kiribati	Framework		8.08	35.2	2310	1965		3.80	3.52	3	Kleypas, GLODAP, World Ocean Atlas 2001, Feely (2009)
Marquesas	Framework		8.08	35.8	2250	1920		4.10	0.75	1	Kleypas, GLODAP, World Ocean Atlas 2001, Feely (2009)
Tahiti	Framework		8.08	36.1	2385	2000		4.40	0.80	1	Kleypas, GLODAP, World Ocean Atlas 2001, Feely (2009)
Tanzania	Framework		8.09	35.0	2310	1980		3.95	1.19	2	Kleypas, GLODAP, World Ocean Atlas 2001, Feely (2009)

Table 2. Average cement abundance and geochemical data per site. N=Total number of coral samples analyzed; TA= Total alkalinity; TCO₂= Total CO₂; pCO₂ =Partial pressure

Puerto Rico contained the second highest cement abundance (15.1%). All samples except PR4 showed significant to extensive amounts of cement. Puerto Rico also boasted the most extensive amount of cement-bearing pores in a single coral (sample PR10 had cement in 41.3% of its intraskeletal pores).

The Japanese samples yielded an average of 10.9% cement. The extent of cementation was quite large ranging from .2% to 26%. The size and clarity of aragonite cement within many of the Japanese samples was often much better than other sites.

All coral samples provided by Dr. Julie Cole (University of Arizona) were grouped together based on their collection method and extremely low amounts of cement. The average cement for all sites was 1.8%. This was unexpected because many of the samples provided by Dr. Cole were collected in locations characterized by moderate to high saturation states (3.6-4.4).

Marine cementation as a function of aragonite saturation states and salinity

The relationship between the average amount of pores containing cement and aragonite saturation state is a scatter plot (Fig. 10) with a very negative correlation that suggests rising cementation with increases ocean acidification. The scatter is generated by the presence of the Key Largo inner reef samples, and the numerous samples from the inside of coral heads provided by Dr. Cole. As explained below in the Discussion, these samples have distinctly different provenance than all others. When they are excluded, the relationship between cement abundance and saturation states improves (Fig. 11A), but scatter still remains, particularly from the presence of more cement in the Galapagos corals relative to the two Panamá sites, and low amounts of cement in the Japan corals and the single Bahamas samples relative to their estimated aragonite saturation states. The relationship between average amount of pores containing cement and

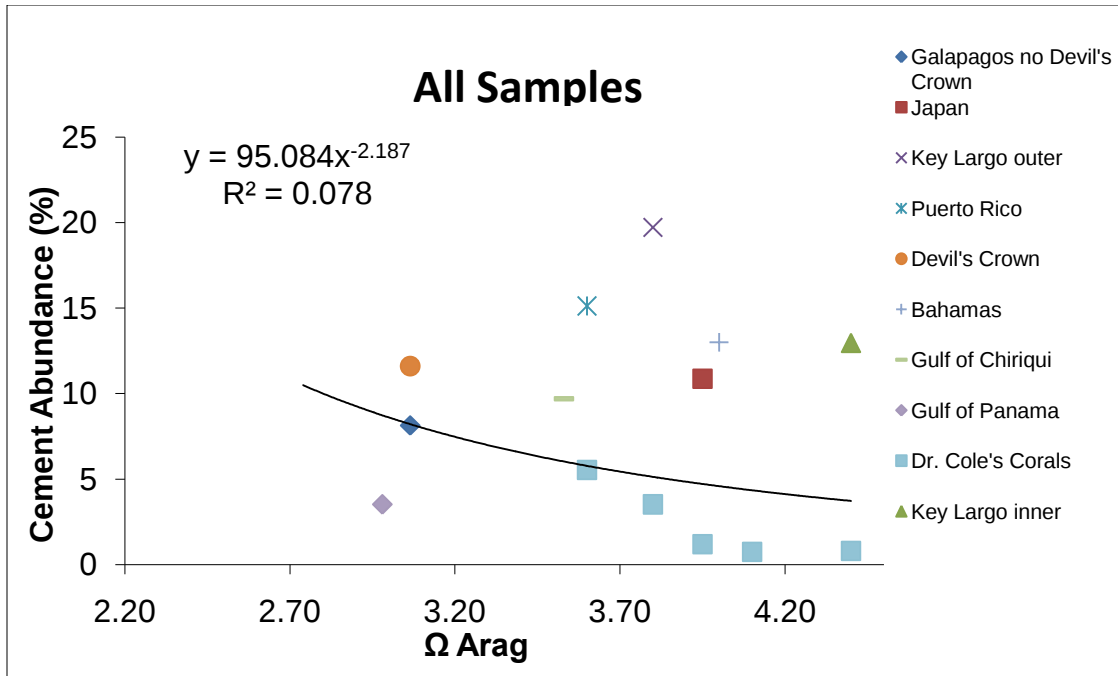


Fig. 10. Power Law relationship between cement abundance and aragonite saturation state for all samples in the study, including samples from the inner shelf of Key Largo and the interior of Pacific coral heads.

aragonite saturation state is strongest when only framework material of similar provenance (i.e., excluding the Cole coral heads) is considered (Fig. 11B). A power law provides the best fit for the aragonite cement and saturation state relationship. This is probably due to the fact that precipitation of both calcite and aragonite follow second order rate laws, meaning precipitation rate is a function of saturation states to a power greater than one (Morse 1983). A similar relationship was explored between the growth rate of gypsum as a function of the relative supersaturation by Hina and Nanocollas (2000). The precipitation rate for gypsum also had an effective reaction order greater than one.

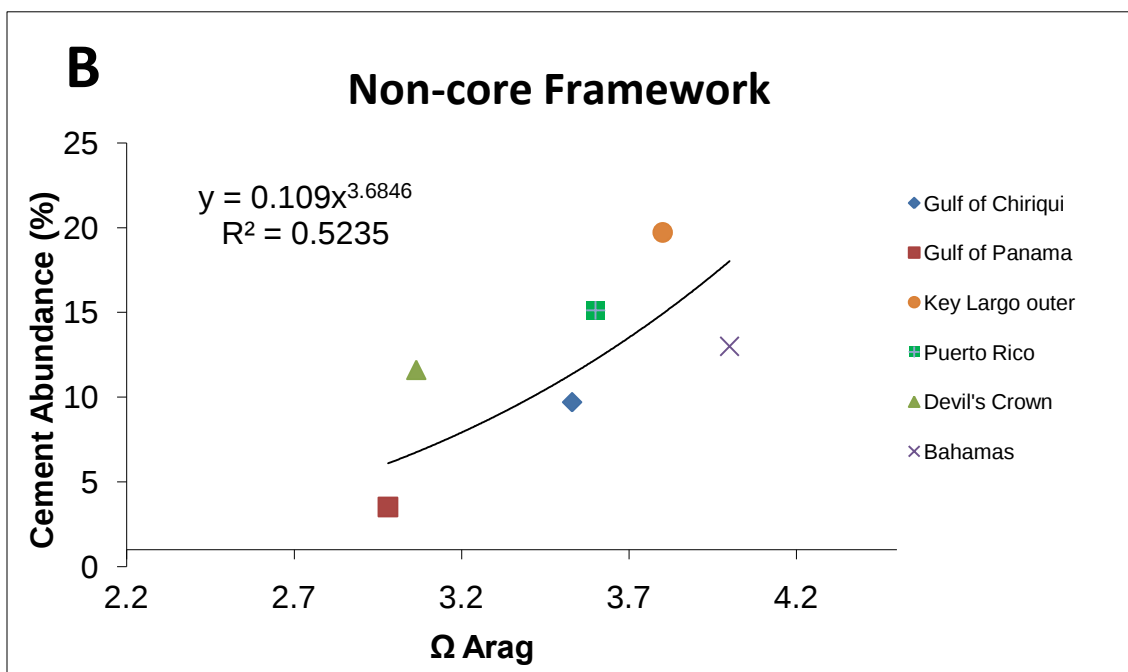
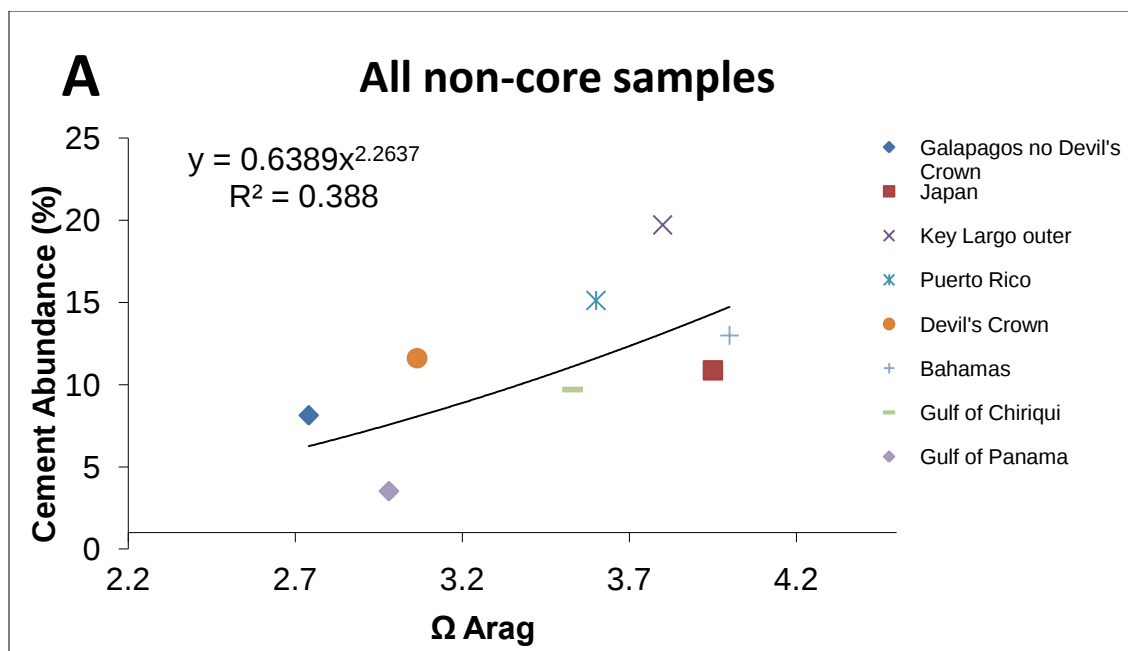


Fig.11. (A) Power law relationships between cement abundance and aragonite saturation states. (A) For all samples (B) Framework samples only.

This study also examined the correlation between cementation and salinity. In comparison to the saturation state relationship, salinity served to be a much better predictor of cement abundance (Fig. 12A). There is also no change between cement and salinity relationship when only framework material was considered (Fig. 12B).

Cement variability in pores

Cement distribution and quantity between intra-skeletal pores is highly variable in all samples (Fig. 13), and cements may or may not occur in pores that also contain detrital sediments or encrusting organisms (Fig. 9). One pore may have cement but an adjacent pore may be empty; conversely cements may be present in most adjacent pores. Similar observations were made by Manzello et al. (2008) and Behairy and El-Sayed (1984). The later authors examined carbonate cements and their distribution throughout El-Qasr reef in the north Red Sea. As in this study Behairy and El-Sayed (1984) found that carbonate cementation varied from pore to pore and that typically not more than one kind of cement was found in any single skeletal void. Yet, adjacent pores can be filled with entirely different materials from sediments to cements, to being completely vacant.

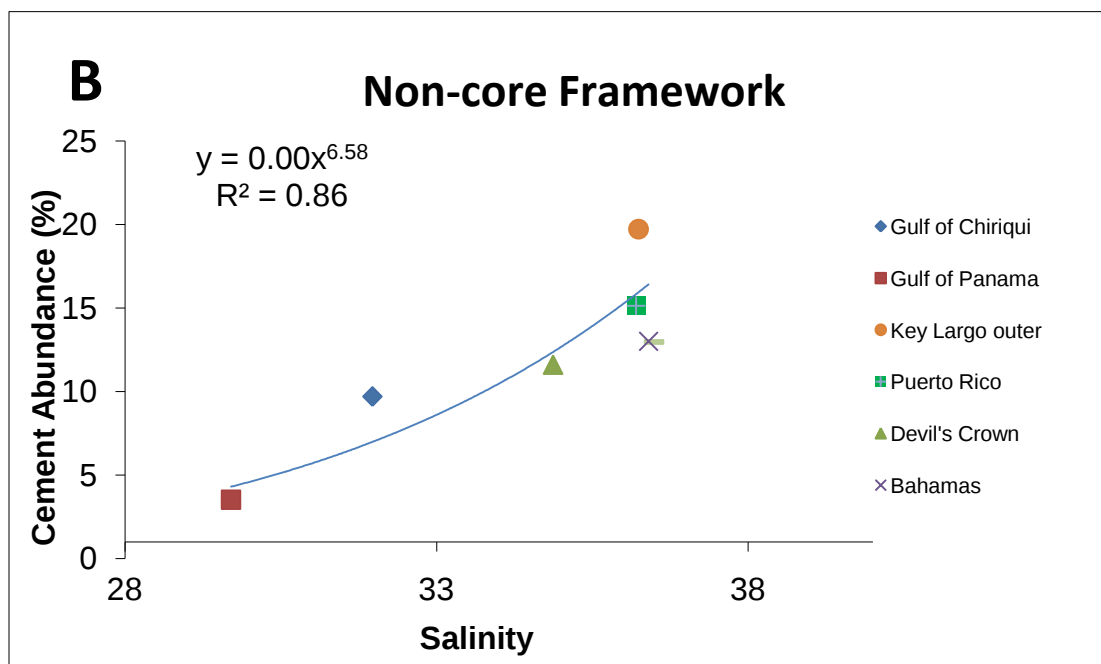
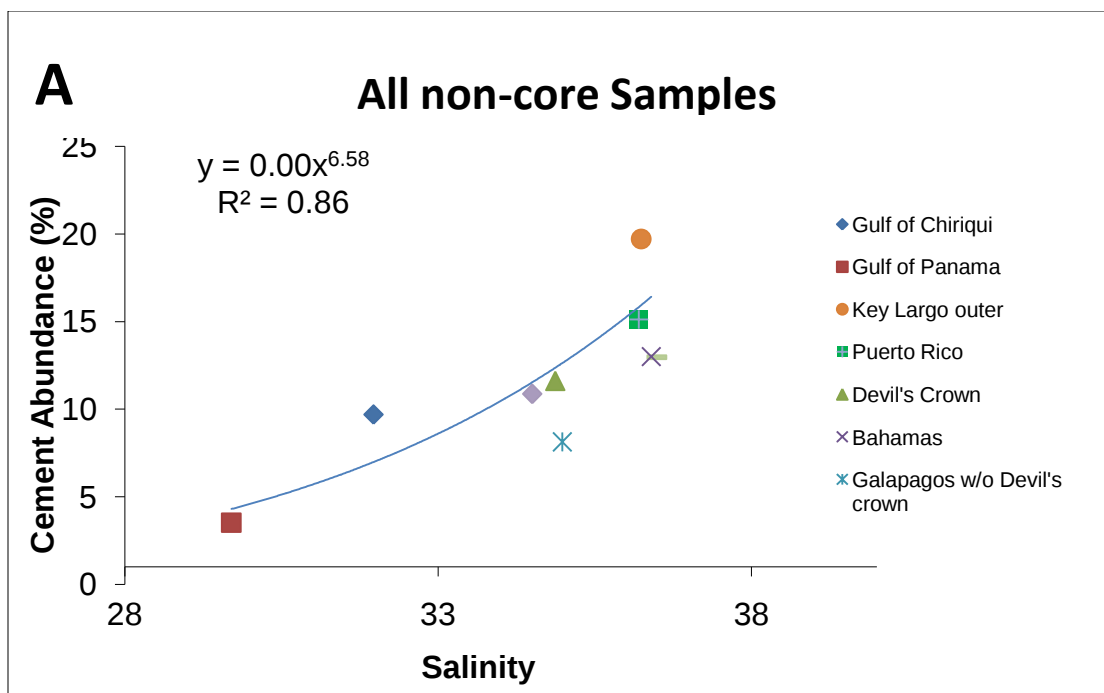


Fig. 12. Relationship between cement abundance and salinity. (A) For all samples (B) Framework samples only.

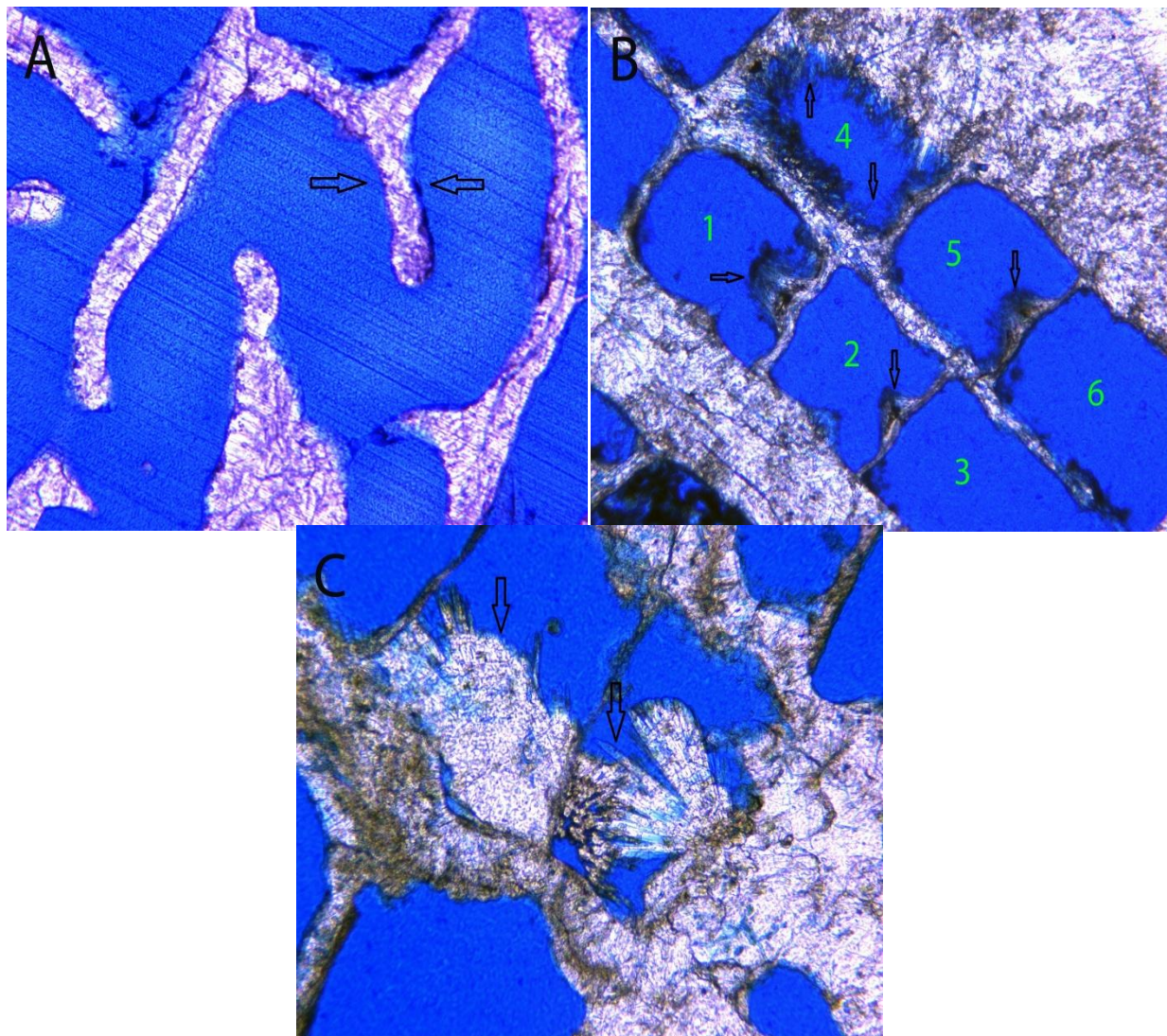


Fig. 13. Thin section photomicrographs of varying cement distributions. All pictures are the same magnification. (A) Sample from Santiago Island, Galápagos with no intra-skeletal cementation. (B) Sample from Key Largo outer shelf with abundant aragonite needles. Cement is present in pores 1,2,4, and 5 (C) An example of botryoidal habit of aragonite crystals (arrows) present in a sample from Okinawa, Japan. Adjacent pores have no cement.

DISCUSSION

The results of this study suggest a correlation between aragonite saturation states and coral reef intra-skeletal cementation, but the correlation is not particularly strong. The best correlation observed, which considered just framework samples from reef surfaces (Fig. 11B), has an R^2 value of only 0.52. This means that only 52% of the variance in cement abundance is explained by aragonite saturation state. The lack of a stronger correlation maybe due to natural variation in cement abundance independent of saturation state, but may also be due to shortcomings in the data base. For almost all samples, aspects of the geochemical data, age restraints on coral, and location of samples within the reef setting were not constant. The potential of these shortcomings is considered below.

Seawater geochemistry on and across reefs is highly variable on a seasonal and annual basis (Crossland 1988; D' Croz and O' Dea 2007; Bates and Amat 2010). This was specifically seen in the Galapagos data where the aragonite saturation state ranged from 2.46 in the dry season to 3.07 in the wet season. Because of this large fluctuation in geochemistry, and the fact that cement abundance is a variable and increases through time, it would be best in a study such as this to use a multi-year average for aragonite saturation. For the Galapagos and Panamá sites average geochemical data was calculated over a period of years throughout both wet and dry seasons, Puerto Rico also has aragonite saturation data that was collected quarterly through a single year. But for all other sites, geochemical data was sampled during one season (Key Largo, FL) or was estimated by online databases such as GLODAP and Ocean Atlas (Japan and Dr. Cole's corals). The later source has the disadvantage of being an extrapolated regional value, not a specific site value. By closely monitoring seawater geochemistry at each sample collection site, this study would have more reliable and consistent data for aragonite saturation states.

Another component of this study that counted on the use of estimates was the age of coral samples, and thus the duration of cementation. This study aimed at using samples that were for ~25-40 years old prior to collection date. The researchers that gathered the materials approximated the ages, so in many cases samples could have been in the water accumulating cement, much longer than similar samples collected at the same location. For example, PR10, which was collected in Puerto Rico, has an abnormally high amount of cement (41%) compared to all other samples. One explanation for the elevated amount of cement is that this sample is older and accumulated cement for a longer period of time in comparison to the rest of the samples collected on the reef crest in Puerto Rico. Given that the high percent of cement in PR10 increases the total average cement of Puerto Rico by 2.2%, it would have been beneficial to have age constraints on all samples collected to ensure a trust worthy data set.

An additional factor that might contribute to scatter in the correlation between saturation states and cement abundance was the location within the reef where samples were collected. As noted in the introduction, different amounts of cementation can be expected in each section of the reef. Corals collected in Puerto Rico were all collected within the same narrow reef crest, whereas corals from Japan varied in collection sites from the reef crest to back reef cobble pavements. In other cases (e.g., Bahamas) a single sample was used as a proxy for an entire reef setting.

Examples of inadequate sample provenance also include the Key Largo inner shelf samples and all of Dr. Cole's samples. The Key Largo inner shelf samples represent a different reefal and geochemical system than all other coral samples. Those shelf samples are from patch reefs formed many kilometers inboard of the shelf margin. In comparison with the Key Largo outer shelf, the inner shelf also had a much higher aragonite saturation state; due to dense sea

grass beds across the shelf taking up ample amounts of CO₂ (Derek Manzello personal communication) . For these reasons all samples from Key Largo inner shelf were excluded from figures 11 and 12.

Coral samples provided by Dr. Julie Cole are also not comparable in their provenance to other samples. Dr. Cole's samples were cored from the center of rapidly growing *Porites* heads and were not in contact with the open ocean waters for the entire 25-40 years of their existence. Once living corals abandoned those pores, the pores were then overgrown by new skeletal mass and isolated from open ocean waters. Isolation from open ocean seawater could explain the lack of aragonite cementation within all samples provided by Dr. Cole. In contrast, all other samples in the study were broken off from coral fragments or collected as reef cobbles, and the pores within these samples were within centimeters of open ocean waters the entire span of their 25-40 years existence.

Gulf of Panamá salinity

Within the salinity-cement relationship (Fig. 10) it is important to be aware of the significance of the Gulf of Panamá. It represents a critical point in the trend line; without it the strength of the correlation (R^2) would decrease substantially. Understanding why the Gulf of Panamá's salinity is so low helps to enhance the understanding of the salinity- cement relationship. The origin of the Panama salinity begins off the coast of Columbia, where the salinity is the lowest in the eastern tropical Pacific due to intense rainfall and river discharge (D' Croz and O' Dea 2007). Large quantities of this low salinity water are brought northward by the Panamá Bright gyre and emptied into the Gulf of Panamá (D' Croz and O' Dea 2007).

Future Cementation in Coral Reefs

Two reef sites in the data base are characterized by seawater aragonite saturation states less than 3.2 and less than 10 % cement (Galapagos excluding devil's crown and the Gulf of Panamá). Both of these locations, because of their low levels of cementation (<10%) in combination with their low aragonite saturation states (<3.2), are at an increased threat of bioerosion and the production of coral debris rather than existing as strong healthy reefs (Manzello et al. 2008). Saturation states at 3 are also considered by Kleypas (2006) to be marginal for reef growth. The data from this study thus suggests that as ocean acidification progresses to the point that all tropical seawaters have low (<3.2) aragonite saturation states, there will be little to no cementation in reefs worldwide and persistent bioerosion will ensue. By 2040, Kleypas (2006) projects that the Earth's oceans will reach these dangerously low saturation states. At that point in time many coral reefs will resemble those of the Galapagos with minimal cementation, heavy bioerosion, and no coral reef framework.

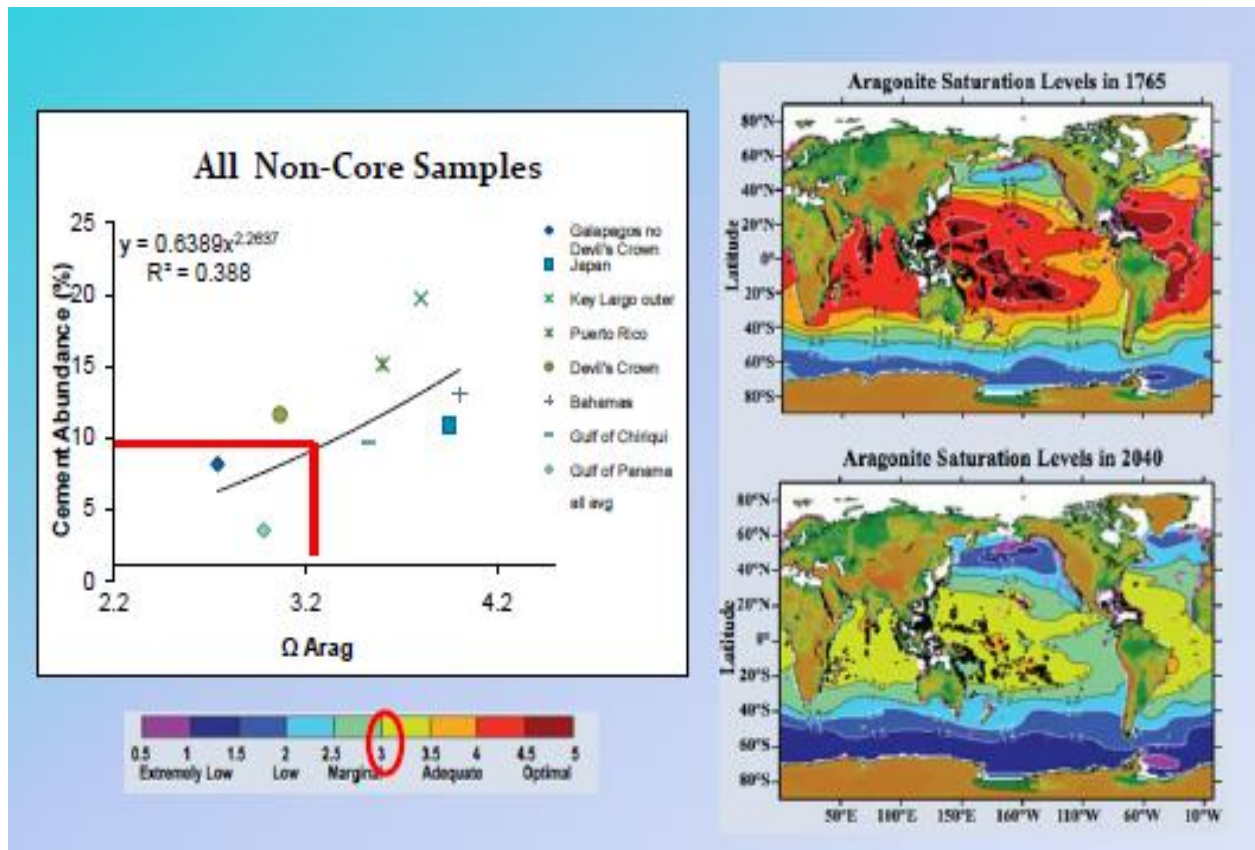


Fig. 14 Model projection of future coral reef cementation. On the right is a figure from Kleypas (2006) showing world ocean aragonite saturation states levels in 1765-2040 with red representing high aragonite saturation states levels and blue and purple representing extremely low aragonite saturation state levels. Aragonite saturation state levels below 3.2 and cement abundance below 10% are characteristic of coral reefs with increased susceptibility to erosion.

CONCLUSION

This study shows that inorganic cement abundance within coral skeletal porosity is related to carbonate ocean chemistry (e.g., saturation state, alkalinity, and $p\text{CO}_2$) on a global scale. As aragonite saturation states continue to decrease in the near future the amount of coral reef cementation will decrease as well. Currently within this study there are two locations where coral reefs are at high risk for erosion due to lack of cementation. If the shortcomings of this study are corrected, the possibility of defining a better empirical relationship between cement

abundance and aragonite saturation state is promising, and it could be a better predictor for future reefs at risk of erosion due to lack of cementation. The findings of this study are just one step in helping to create a model that will be an accurate predictor for future coral reef cementation in a high CO₂ world with ocean pH at historic lows.

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Appendix 1. Seawater geochemistry and cement abundance for individual sample sites.

Sample	Location	Reef name/ Island	Reef framework	Location in reef	pH	Salinity	TA	TCO2	pCO2	Arag. Saturation state	Cement Abundance
KLI-1	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	5.4
KLI-2	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	24.1
KLI-3	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	2.0
KLI-4	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	11.4
KLI-5	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	4.7
KLI-6	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	16.2
KLI-7	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	28.1
KLI-8	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	16.7
KLI-10	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	7.8
KLI-11	Key Largo-inner		framework		8.17	36.5	2327	1925	27.2	4.40	13.2
KLO-1	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	22.5
KLO-2	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	49.4
KLO-3	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	28.3
KLO-4	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	30.7
KLO-5	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	4.2
KLO-6	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	8.1
KLO-7	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	17.5
KLO-8	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	14.6
KLO-9	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	5.1
KLO-10	Key Largo-outer		framework		8.07	36.2	2386	2045	37.8	3.80	16.8
PR1	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	16.7
PR2	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	12.3
PR3	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	4.8
PR4	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	1.0
PR5	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	14.0
PR6	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	19.8
PR7	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	24.2
PR8	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	23.6
PR9	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	6.0
PR10	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	41.3
PR11	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	7.0
PR12	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	14.1
PR13	Puerto Rico	Cayo Enrique, La Paraguera	framework	fore-reef		36.2				3.60	11.7
DC2b-Rc	Galapagos Is.	Devil's Crown, Floreana Is.	framework		7.98	34.9	2316	2073	47.9	3.06	2.0
DC1-1A	Galapagos Is.	Devil's Crown, Floreana Is.	framework		7.98	34.9	2316	2073	47.9	3.06	3.9
DC3c-Rc	Galapagos Is.	Devil's Crown, Floreana Is.	framework		7.98	34.9	2316	2073	47.9	3.06	4.1
DC1-Rc	Galapagos Is.	Devil's Crown, Floreana Is.	framework		7.98	34.9	2316	2073	47.9	3.06	16.2
DC2c-Rc	Galapagos Is.	Devil's Crown, Floreana Is.	framework		7.98	34.9	2316	2073	47.9	3.06	19.3
DC2a-Rc	Galapagos Is.	Devil's Crown, Floreana Is.	framework		7.98	34.9	2316	2073	47.9	3.06	24.1

B1-A	Galapagos Is.	Bartolome, Santiago Is.	NOT		7.91	35.0	2309	2073	58.5	2.74	0.0
BART2-Rc	Galapagos Is.	Bartolome, Santiago Is.	NOT	reef crest	7.91	35.0	2309	2073	58.5	2.74	5.0
BART1-Rc	Galapagos Is.	Bartolome, Santiago Is.	NOT	reef crest	7.91	35.0	2309	2073	58.5	2.74	6.6
SCST B3	Galapagos Is.	San Cristobal	NOT		7.91	35.0	2309	2073	58.5	2.74	8.3
SCST B1	Galapagos Is.	San Cristobal	NOT		7.91	35.0	2309	2073	58.5	2.74	18.2
SF2-2	Galapagos Is.	Santa Fe	NOT		7.91	35.0	2309	2073	58.5	2.74	0.0
SF2-3	Galapagos Is.	Santa Fe	NOT		7.91	35.0	2309	2073	58.5	2.74	2.8
SF 1b-Rc	Galapagos Is.	Santa Fe I.	NOT	reef crest	7.91	35.0	2309	2073	58.5	2.74	5.8
SF 1a-Rc	Galapagos Is.	Santa Fe I.	NOT	reef crest	7.91	35.0	2309	2073	58.5	2.74	7.0
DBS-Rc			NOT	reef crest	7.91	35.0	2309	2073	58.5	2.74	12.7
SF1-4	Galapagos Is.	Santa Fe	NOT		7.91	35.0	2309	2073	58.5	2.74	23.1
LSI-1	Bahamas	Lee Stocking I.	framework		8.07	36.4	2382	2028	37.2	4.00	13.0
SEC-FR6	Panamá - G. of Chiriquí	Secas Is.	framework	fore reef	8.04	32.0	2093	1797	41	3.53	1.6
SEC-RF2	Panamá - G. of Chiriquí	Secas Is.	framework	reef flat	8.04	32.0	2093	1797	41	3.53	3.4
SEC-RF1	Panamá - G. of Chiriquí	Secas Is.	framework	reef flat	8.04	32.0	2093	1797	41	3.53	3.8
SEC-RF8	Panamá - G. of Chiriquí	Secas Is.	framework	reef flat	8.04	32.0	2093	1797	41	3.53	4.6
SEC-FR5	Panamá - G. of Chiriquí	Secas Is.	framework	fore reef	8.04	32.0	2093	1797	41	3.53	4.9
SEC-FR4	Panamá - G. of Chiriquí	Secas Is.	framework	fore reef	8.04	32.0	2093	1797	41	3.53	9.3
SEC-FR3	Panamá - G. of Chiriquí	Secas Is.	framework	fore reef	8.04	32.0	2093	1797	41	3.53	13.2
SEC-RF7	Panamá - G. of Chiriquí	Secas Is.	framework	reef flat	8.04	32.0	2093	1797	41	3.53	21.7
UVA-FR8	Panamá - G. of Chiriquí	Uva Reef	framework	fore reef	8.01	32.0	2093	1797	41	3.53	1.9
UVA-RB4	Panamá - G. of Chiriquí	Uva Reef	framework	reef base	8.01	32.0	2093	1797	41	3.53	2.3
UVA-RB1	Panamá - G. of Chiriquí	Uva Reef	framework	reef base	8.01	32.0	2093	1797	41	3.53	3.6
UVA-BR1	Panamá - G. of Chiriquí	Uva Reef	framework	back reef	8.01	32.0	2093	1797	41	3.53	9.0
UVA-FR6	Panamá - G. of Chiriquí	Uva Reef	framework	fore reef	8.01	32.0	2093	1797	41	3.53	9.6
UVA-FR5	Panamá - G. of Chiriquí	Uva Reef	framework	fore reef	8.01	32.0	2093	1797	41	3.53	11.8
UVA-RF9	Panamá - G. of Chiriquí	Uva Reef	framework	fore reef	8.01	32.0	2093	1797	41	3.53	13.4
UVA-RB5	Panamá - G. of Chiriquí	Uva Reef	framework	reef base	8.01	32.0	2093	1797	41	3.53	17.1
UVA-RF1	Panamá - G. of Chiriquí	Uva Reef	framework	reef flat	8.01	32.0	2093	1797	41	3.53	20.9
UVA-RF5	Panamá - G. of Chiriquí	Uva Reef	framework	reef flat	8.01	32.0	2093	1797	41	3.53	22.5
SAB-RF4	Panamá- G. of Panamá	Saboga Reef	framework	reef flat	8.04	29.7	1969	1717	36.7	2.98	1.5
SAB-FR5	Panamá- G. of Panamá	Saboga Reef	framework	fore reef	8.04	29.7	1969	1717	36.7	2.98	1.7
SAB-RF1	Panamá- G. of Panamá	Saboga Reef	framework	reef flat	8.04	29.7	1969	1717	36.7	2.98	1.8
SAB-FR3	Panamá- G. of Panamá	Saboga Reef	framework	fore reef	8.04	29.7	1969	1717	36.7	2.98	1.9
SAB-FR6	Panamá- G. of Panamá	Saboga Reef	framework	fore reef	8.04	29.7	1969	1717	36.7	2.98	2.6
SAB-RF7	Panamá- G. of Panamá	Saboga Reef	framework	reef flat	8.04	29.7	1969	1717	36.7	2.98	11.6

A-5	SE Saipan Island	Laolao Bay	NOT	reef edge (fringing reef)	8.11	34.6	2275	1965		3.95	0.2
A-4	SE Saipan Island	Laolao Bay	NOT	reef edge (fringing reef)	8.11	34.6	2275	1965		3.95	1.0
A-3	NW Guam Island	Double reef	NOT	in reef (fringing reef)	8.11	34.5	2250	1900		3.95	1.4
Ikei-1	Okinawa, Japan	Ikei-jima reef	NOT		8.11	34.5	2295	1960		3.95	1.5
A-1	Kodakara-jima, Japan		NOT	reef crest	8.11	34.6	2295	1960		3.95	2.2
A-2	Kodakara-jima, Japan		NOT	reef crest	8.11	34.6	2295	1960		3.95	2.3
M-1B	Okinawa, Japan	Maeda	NOT	Fore- reef	8.11	34.5	2295	1960		3.95	6.7
Ikei-2	Okinawa, Japan	Ikei-jima reef	NOT	reef crest	8.11	34.5	2295	1960		3.95	15.5
Ikei-3	Okinawa, Japan	Ikei-jima reef	NOT	reef crest	8.11	34.5	2295	1960		3.95	23.2
Ikei-4	Okinawa, Japan	Ikei-jima reef	NOT	reef crest	8.11	34.5	2295	1960		3.95	24.7
M-3	Ishigaki Is., Japan	Kabira	NOT		8.11	34.1	2295	1960		3.95	25.8
M-1A	Okinawa, Japan	Maeda	NOT	Fore- reef	8.11	34.5	2295	1960		3.95	26.0
GBR-050	Australia	Great Barrier Reef		outer shelf		35.3	2330	1975		3.60	5.5
ARA-95	Kiribati	Aranuka			8.08	35.2	2310	1965		3.80	0.3
Mai2-3	Kiribati	Maiana			8.08	35.2	2310	1965		3.80	4.6
ONO-6	Kiribati	Onotoa			8.08	35.2	2310	1965		3.80	0.2
NH6-2a	Marquesas	nukuhiva			8.08	35.8	2250	1920		4.10	0.7
Tahiti	Tahiti				8.08	36.1	2385	2000		4.40	0.8
Pem-4B2	Tanzania	Pemba Is.			8.09	35.0	2310	1980		3.95	0.5
Z1B1	Tanzania	Zanzibar Is.			8.09	35.0	2310	1980		3.95	1.9