Ocean Acidification has Impacted Coral Growth on the Great Barrier Reef

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Key Points:

- Numerical model of coral growth isolates the respective impacts of ocean acidification and ocean warming on coral growth.
- Ocean acidification has caused ~13% decline in the skeletal density of massive *Porites* corals on the Great Barrier Reef since 1950.
- OA-induced thinning of coral skeletons reflects enhanced acidification of reef water relative to the surrounding open ocean.

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Abstract

Ocean acidification (OA) reduces the concentration of seawater carbonate ions that stony corals need to produce their calcium carbonate skeletons, and is considered a significant threat to the functional integrity of coral reef ecosystems. However, detection and attribution of OA impact on corals in nature are confounded by concurrent environmental changes, including ocean warming. Here we use a numerical model to isolate the effects of OA and temperature, and show that OA alone has caused 13±3% decline in the skeletal density of massive *Porites* corals on the Great Barrier Reef since 1950. This OA-induced thinning of coral skeletons, also evident in *Porites* from the South China Sea but not in the central equatorial Pacific, reflects enhanced acidification of reef water relative to the surrounding open ocean. Our finding reinforces concerns that even corals that might survive multiple heatwaves are structurally weakened and increasingly vulnerable to the compounding effects of climate change.

1. Introduction

About 1/3 of the CO_2 emitted to the atmosphere by human activities has been absorbed by the oceans, driving about 0.1 unit decline in ocean pH and a corresponding ~20% decrease in carbonate ion concentration ($[CO_3^{2-}]$) since the pre-industrial era (e.g., Doney et al., 2009; Feely et al., 2009; Friedlingstein et al., 2019). This process, known as ocean acidification (OA), is expected to continue through this century and beyond, causing another 0.1-0.4 unit pH decline by 2100 and effectively halving the concentration of carbonate ions in seawater relative to the pre-industrial era (e.g., Orr et al., 2005; Doney et al., 2009; Feely et al., 2009). Calcifying organisms, which need carbonate ions to form their skeletons, are most at risk and coral reef ecosystems are expected to be heavily impacted (e.g., Orr et al., 2005; Hoegh-Guldberg et al., 2007). Laboratory experiments that reared corals and other coral reef calcifiers under high CO_2 conditions, as well as field studies of naturally low-pH reefs indicate, in general, that decreased rates of calcification and increased rates of dissolution and bioerosion have, and will continue to occur, as the tropical oceans become more acidified over the next few decades (e.g., Pandolfi et al., 2011; Chan and Connolly, 2013).

Nevertheless, while measurable ocean acidification of the tropical ocean has been underway for several decades now, detection and attribution of the effects of ocean acidification on reef-building corals have been challenging. Century-long records of coral calcification rates generated from skeletal cores do not show a consistent decline in calcification rates as ocean pH decreased through the 20th century. Rather, in some locations, coral calcification rates sharply decreased, others remained stable, and yet others increased over this time period (e.g., Cooper et al., 2008; De'ath et al., 2009; Cooper et al., 2012; D'Olivo et al., 2013). Even where declines in calcification have been observed, our ability to attribute such changes to ocean acidification is

confounded by the fact that ocean warming, sea level rise, changes in surface ocean productivity, as well as many localized anthropogenic disturbances, are co-occurring with ocean acidification and also influence coral growth (e.g., Cooper et al., 2008; De'ath et al., 2009; Lough and Cantin, 2014; Pandolfi, 2015).

Massive long-lived *Porites* colonies are common on reefs across the Indo-Pacific, and their growth histories span the time period over which ocean acidification has occurred, providing a unique continuous archive of such impacts. The skeletal growth of *Porites* corals occurs in two steps in which the corals initially extend existing skeletal elements to enable upward growth, i.e., extension, followed by thickening of those elements, i.e., densification (Barnes and Lough, 1993). Extension, which is driven primarily by the creation of calcification "centers" or nucleation sites, is under strong biological control (presumably through the organic matrix) and less sensitive to ocean acidification (e.g., Cohen and McConnaughey, 2003; Nothdurft and Webb, 2007; Crook et al., 2013; Fantazzini et al., 2015; Tambutte et al., 2015; Mollica et al., 2018). In contrast, densification, which contributes the bulk of the skeletal mass and serves to reinforce the skeleton against the force of the waves and currents, is strongly sensitive to ocean acidification because crystal formation during this phase is under strong physicochemical control (e.g., Crook et al., 2013; Fantazzini et al., 2015; Tambutte et al., 2015; Mollica et al., 2018; Rippe et al., 2018; Martinez et al., 2019). Here we compile existing and new skeletal growth records of 95 *Porites* corals from the Great Barrier Reef (De'ath et al., 2009), South China Sea (Su et al., 2016) and the central equatorial Pacific Ocean spanning the time period of 1871 to 2014 (Methods, Fig. S1), and use a coral skeletal growth model to isolate the effects of different factors and quantify their respective contributions to the coral growth, particularly to coral skeletal density.

2. Methods

2.1 Porites skeletal growth parameters

Great Barrier Reef (GBR) and South China Sea (SCS, Hainan Island) reefs: Our analysis employs the annual *Porites* skeletal growth parameters (i.e., extension, density and calcification) reported in previous studies of the Great Barrier Reef (De'ath et al., 2009) and the South China Sea (Hainan Island) reefs (Su et al., 2016). For the Great Barrier Reef, an updated version of the skeletal growth parameters originally reported in (De'ath et al., 2009) is adopted, which excludes the incomplete outmost growth layers (De'ath et al., 2013). Although some other studies also reported *Porites* skeletal growth parameters on the Great Barrier Reef (e.g., D'Olivo et al., 2013), those data are not available in online repositories and are thus not included in our analysis. To robustly investigate the multi-decadal variations in *Porites* skeletal growth, we focus on the time periods when at least 10 skeletal cores are available for each year and include only cores that have at least 50 years of growth within these selected time periods. Furthermore, we exclude the time periods when independent constraints of environmental parameters (e.g., temperature, Rayner et al., 2003) are not available. Together, these selection criteria result in the inclusion of 60 *Porites* cores from 39 reefs over the period of 1871-2000 for the Great Barrier Reef, and 16 *Porites* cores from 2 reefs over the period of 1901-2000 for the Hainan Island, South China Sea (Fig. S1). For each core, we calculate the percentage changes in its annual extension, density and calcification relative to the corresponding mean values over 1951-1960, a common period that all cores cover (Fig. S2, Data S1): $x_{i,rel} = \left(x_i / \overline{x_{i,c}} - 1 \right) \times 100$, where x_i is the skeletal growth parameter at a given year i and $\overline{x_{i,c}}$ is the mean value of that parameter over the common period for the same core.

Central Equatorial Pacific (CEP) reefs: *Porites* skeletal cores were collected from 7 central equatorial Pacific reefs (Jarvis, Kanton, Kingman, Kiritimati, Nikumuroro, Rawaki, Tutuila), and were imaged with a Siemens Volume Zoom Spiral Computerized Tomography scanner. Annual extension rates, skeletal densities and calcification rates were then determined based on these CT images along polyp growth axes (Data S2, DeCarlo et al., 2015; Mollica et al., 2018). Similar to the GBR and South China Sea reef cores, we focus on the time periods when at least 10 skeletal cores are available for each year and include only cores that have at least 10 years of growth within these selected time periods. These lead to the inclusion of 19 *Porites* cores over the period of 1978-2014 in our analysis (Fig. S1). We then calculate, for each core, the percentage changes in its annual extension, density and calcification relative to the corresponding mean values over 1998-2007, a common period that all the CEP coral cores cover (see above; Fig. S2, Data S1).

2.2 Predicting *Porites* skeletal density with a skeletal growth model

For each of our selected *Porites* cores, we predict its annual skeletal densities based on a skeletal growth model (Mollica et al., 2018, Text S1) and use them to evaluate and isolate the effects of three main factors that influence *Porites* skeletal density, i.e., extension, temperature, and seawater carbonate chemistry. This skeletal growth model, building on previous studies of *Porites* skeletal growth (Barnes and Lough, 1993; Taylor et al., 1993), explicitly simulates the two distinct phases of *Porites* skeletal growth (i.e., extension and densification) and has quantitatively reproduced the experimentally measured *Porites* skeletal densities from a variety of reef environments (Mollica et al., 2018).

For each core, model simulations were conducted under three different conditions: (1) extension as the only forcing, i.e., keeping temperature and seawater carbonate chemistry

constant; (2) temperature as the only forcing, i.e., keeping extension and seawater carbonate chemistry constant; (3) extension and temperature combined as the forcing, i.e., keeping seawater carbonate chemistry constant. For the parameters which were kept constant during the model simulations, their values were set as the mean values over the common periods for each core, i.e., 1951-1960 for the Great Barrier Reef and the South China Sea reef cores, and 1998-2007 for the central equatorial Pacific reef cores.

For all the model simulations, experimentally measured extension, seawater temperatures from the Hadley Centre Sea Ice and Sea Surface Temperature (HadiSST) v1.1 dataset (1° × 1°, Rayner et al., 2003), and seawater pH and DIC outputs from the Community Earth System Model Biogeochemical historical run (CESM-BGC, Hurrell et al. 2013) were used. HadiSST v1.1 dataset, although relatively coarse in spatial resolution compared to some other temperature datasets (e.g., OISST, Banzon et al., 2016), covers the whole time period of our core records (i.e., from 1871 to 2014) and has been used extensively in previous reef studies (e.g., De'ath et al., 2009). Similarly, seawater pH and DIC outputs from the CESM-BGC historical run were adopted due to the lack of constraints on past seawater carbonate chemistry in most reefs; but since seawater pH and DIC were kept constant in all our model simulations (see above), their exact values have negligible effects on our results which focus on the relative percentage changes in skeletal density (Mollica et al., 2018). Other parameters in the *Porites* skeletal growth model (e.g., corallite area, tissue thickness) were assumed to be constant over the growth of each coral and set as either the average values reported for *Porites* corals in the region or the optimized values derived from *Porites* corals from multiple tropical reefs (Table S1, Lough and Barnes, 2000; Su et al., 2016; Mollica et al., 2018). We then analyzed these model predicted skeletal densities the same way as the experimentally measured densities, and calculated, for

each core, the percentage changes in the model predicted annual densities relative to the corresponding mean of the common periods.

2.3 Constraints on the reef-water pH on the Great Barrier Reef

We compiled existing constraints on the reef-water pH on the Great Barrier Reef based on the boron isotope composition of coral skeletons at six GBR reef sites, including Arlington Reef, Flinders Reef, Pandora Reef, Havannah Island, Rib Reef and Reef 17-065 (Pelejero et al., 2005; Wei et al., 2009; D'Olivo et al., 2015). To robustly investigate the multi-decadal variations in reef-water pH at the Great Barrier Reef, we focused on the time periods when at least 5 pH records were available for each year. This limited our analysis to the period from 1966 to 2000 for the Great Barrier Reef (Fig. S3, 7 *Porites* cores in total). To remove the methodological inconsistency in pH estimations among different studies (e.g., regarding the use of boron isotope fractionation factors), we recalculated the seawater pH values based on the coral $\delta^{11}B$ data reported in each study (Trotter et al., 2011; D'Olivo et al., 2015) using the same physicochemical parameters (Text S2, Data S3, Dickson, 1990; Klochko et al., 2006; Foster et al., 2010). Then, for each $\delta^{11}B_{coral}$ record, we calculated changes in its estimated pH_{sw} relative to the corresponding mean over 1976-1985, a common period that all these records cover (Fig. S3, Data S3).

There are currently very limited constraints on the reef-water pH for the South China Sea and the central equatorial Pacific reefs (two for the South China Sea reefs, Liu et al., 2014; Wei et al., 2015; none for the central equatorial Pacific reefs), which precludes robust statistical analysis of the temporal trends in reef-water pH for these two regions.

2.4 Determining temporal trends in *Porites* skeletal growth parameters and reef-water pH

We determined the temporal trends in (1) *Porites* skeletal growth parameters (both experimentally measured and model predicted) on the Great Barrier Reef, the South China Sea and the central equatorial Pacific and (2) the reef-water pH records we compiled and recalculated for the Great Barrier Reef, using the Generalized Additive Mixed Models (GAMMs). GAMMs are extensions to the Generalized Linear Mixed Models (GLMMs) and allow for non-linear response (Wood, 2017). Similar methods have been used in previous studies of *Porites* skeletal growth (e.g., De'ath et al., 2009; Cooper et al., 2012; De'ath et al., 2013; Ridd et al., 2013). In each model, year was set as the fixed effect component, while coral colony and reef site as the random effects components. A smoothing spline was applied to the fixed effect variable, with the degree of smoothness (i.e., the degrees of freedom associated with the smoothing function) determined through cross-validation (Cooper et al., 2012). All the models were analyzed using the *lme4* and *mgcv* packages in the R programming software with the REML method (Wood, 2011; Bates et al., 2015).

3. Results and Discussion

Temporal trends in skeletal growth parameters, determined using generalized additive mixed models (Methods, Wood, 2017), show that *Porites* skeletal density has decreased by ~11% on the Great Barrier Reef from 1871 to 2000 (Fig. 1). This density decline is most significant between 1980 and 2000 reaching ~3% per decade, compared to ~0.5% per decade from 1871 to 1980. Declines in skeletal density are also evident in *Porites* colonies in the South China Sea and in the central equatorial Pacific, ranging from ~20% over 1901-2000 to ~7% over 1978-2014, respectively (Fig. S4).

These density declines, although consistent with the negative impacts expected from OA, cannot be attributed exclusively to OA, because factors including skeletal extension (E) and seawater temperature also influence coral skeletal density (Fig. 2). Specifically, as extension increases, density will decrease, even if ocean pH does not change, because the less time a skeletal element resides within the coral tissue layer (i.e., at higher extension rate) the less it can be thickened, and vice versa (Barnes and Lough, 1993; Taylor et al., 1993; Mollica et al., 2018). Conversely, skeletal density is expected to increase as seawater temperature increases, because calcium carbonate precipitates faster at higher temperatures (Burton and Walter, 1987; Mollica et al., 2018). Note, other environmental factors such as nutrients and water quality can also affect coral growth mostly through their influences on extension (e.g., Dustan, 1975; Huston, 1985; Tomascik and Sander, 1985; Lough and Cooper, 2011; Al-Rousan, 2012), and are thus implicitly included in our analysis as well (Fig. 2).

Indeed, at all three study sites, the density declines observed in the *Porites* corals over time were accompanied by increases in skeletal extension, ranging from ~9% at the Great Barrier Reef to ~30% in the South China Sea, and by temperature variations up to 1.5^{o}C (Fig. 1, 3, S4-S5). We estimated the contribution of these extension and temperature changes to the observed density declines, using a skeletal growth model that explicitly simulates the two-step skeletal growth of *Porites* corals and links the coral calcifying fluid chemistry with external seawater conditions (Methods, Mollica et al., 2018). The model predicts that skeletal extension acting alone would have caused a modest decline in skeletal density of ~4% on the Great Barrier Reef over the time period from 1871 to 2000 (Fig. 3a). Conversely, variations in water temperature alone, especially the increase post-1950, would have caused a ~6% increase in skeletal density over the same time period (Fig. 3b). Together, skeletal extension and ocean warming are

expected to lead to oscillations of *Porites* skeletal density between -2% and 2% during 1871-2000, not the decrease in density as observed (Fig. 3c).

Ocean acidification, skeletal extension and temperature are the main influences of the skeletal density of *Porites* corals (Fig. 2, Mollica et al., 2018). Previous model simulations, explicitly considering these three factors, have quantitatively reproduced the experimentally measured *Porites* skeletal densities from a variety of reef environments (Mollica et al., 2018). Thus, having constrained the effects of temperature and extension, we can assess the impact of OA on *Porites* density (ΔD_{OA}) by subtracting the model predicted density changes induced by extension and temperature variations ($\Delta D_{E+T,\ modeled}$) from the measured density changes ($\Delta D_{measured}$):

$$\Delta D_{OA} = \Delta D_{measured} - \Delta D_{E+T,\ modeled} \quad (1)$$

Figure 3c shows that the OA-driven changes in the skeletal density of *Porites* corals on the Great Barrier Reef since 1871. OA-driven changes are small, fluctuating between 2 and -2%, until 1950, after which a rapid ~13±3% (95% confidence interval) decline in density is observed from 1950 to 2000. The timing of the OA effect is consistent with the accelerated decline in the ocean pH post-1950. However, the magnitude of the ocean pH decrease post-1950 is too small to explain the dramatic decline in skeletal density on the GBR. In fact, the skeletal growth model predicts a density change of ~1% given a 0.06 change in seawater pH alone.

This discrepancy is explained by the enhanced acidification of reef-water on the GBR relative to the open ocean (Fig. 4a, Methods, Pelejero et al., 2005; Wei et al., 2009; D'Olivo et al., 2015). In particular, boron isotope analysis of *Porites* skeleton shows that reef-water pH on the

Arlington Reef (mid-shelf, northern GBR) has decreased by ~0.2 unit since the 1940s (Methods, Wei et al., 2009), which is about 2.5-fold larger than the decrease in open ocean pH over the same time period (i.e., ~0.06 unit). This enhanced acidification of reef-water has also been observed at inshore reefs in the GBR and at other coral reefs around the globe, where reef-water pCO_2 has increased 2.5~3.5-fold faster than the open ocean over the past 20-30 years potentially as a result of increased inputs of terrestrial nutrient and organic matter (Fig. 4a, Cyronak et al., 2014; Uthicke et al., 2014).

Our model does predict some high-frequency density variability that is not replicated in the measured data, especially around 1940. This variability is driven by the temperature input to the model. It is likely that well-documented uncertainties in the historical temperature data-products during this time period (e.g., Chelton and Risien, 2016) confound our estimation of the temperature effects on *Porites* density and caused the ~4% decline in density around 1940 that we currently attribute to OA. Efforts to increase the accuracy of the historical temperature records (e.g., Chan et al., 2019) will improve our estimates of the OA impact on coral skeletal growth. Overall however, the model-predicted effects of extension and temperature on skeletal density are in good agreement with the independently measured density changes (Fig. 3a-b) and our results indicate strong impacts of OA on GBR corals post-1950.

Analysis of *Porites* growth records from Hainan Island, South China Sea indicate that here too, OA has caused ~7±3% (95% confidence interval) decline in *Porites* skeletal density from 1901 to 2000, with the most significant decline also starting around 1950 (Fig. 4b, S5, Text S4). The similar timing of the estimated OA impacts on the GBR and the South China Sea corals occurs despite the dramatically different bulk skeletal growth records at these two regions (Fig. S4), and is consistent with the similar evolution of ocean pH around the GBR and the South

China Sea (Fig. 4b). In contrast, analysis of central equatorial Pacific reef corals suggests that OA has not yet had a significant influence on *Porites* growth in the region (Fig. 4b, S5, Text S4). This is likely because these reefs are bathed in open-ocean seawaters and have thus experienced relatively modest decreases in their reef-water pH to date.

Our study presents strong evidence that 20th century ocean acidification, exacerbated by reef biogeochemical processes, has had measurable effects on the growth of a key reef-building coral across the Great Barrier Reef and in the South China Sea, and will likely accelerate as ocean acidification progresses over the next several decades. While it is difficult to directly extrapolate our estimated OA impact to any specific reef due to the variability of reef-water pH among and within different reefs (e.g., Gagliano et al., 2010; Uthicke et al., 2014; D'Olivo et al., 2015; Mongin et al., 2016), we expect broadly similar magnitudes of OA impact worldwide because enhanced acidification of reef water pH similar to the GBR has been observed in many coral reefs around the globe (Fig. 4a, Cyronak et al., 2014).

Declines in coral skeletal density increase the susceptibility of coral skeletons to bioerosion, dissolution and storm damage (e.g., Madin et al., 2012; Wisshak et al., 2012; Crook et al., 2013; DeCarlo et al., 2015; Fantazzini et al., 2015), and suggest structurally weaker and more vulnerable coral reefs in the 21st century. In particular, the strength of coral skeleton decreases exponentially with decreasing skeletal density, e.g., about 60% reduction of compressive strength for a 13% density decline (Chamberlain, 1978; Scott and Risk, 1988; Madin, 2004; Madin et al., 2008), making the coral skeletons increasingly susceptible to storm damage. This, together with the deleterious influences of other global and local environmental stressors (e.g., ocean warming, sea level rise, pollution), poses severe challenges for the health and survival of coral reef ecosystems and their exceptional biodiversity. Better understanding of

the controls on the reef-water pH and coral calcification mechanisms will enable more accurate projections of OA impacts on coral reef ecosystems and thus the developments of potential mitigation strategies.

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The data generated in this study are being archived at the NOAA National Centers for Environmental Information-Paleoclimatology Data repository, i.e., the World Data Service for Paleoclimatology. For review purposes, a copy of the data has been provided in the Supporting Information (Data S1-S3).

References

Al-Rousan, S. (2012) Skeletal extension rate of the reef building coral Porites species from Aqaba and their environmental variables. *Nat. Sci.* 4, 731-739.

Banzon, V., Smith, T.M., Chin, T.M., Liu, C.Y. and Hankins, W. (2016) A long-term record of blended satellite and in situ sea-surface temperature for climate monitoring, modeling and environmental studies. *Earth System Science Data* 8, 165-176. https://doi.org/10.5194/essd-8-165-2016

Barnes, D.J. and Lough, J.M. (1993) On the nature and causes of density banding in massive coral skeletons. *J. Exp. Mar. Biol. Ecol.* 167, 91-108. https://doi.org/10.1016/0022-0981(93)90186-r

Bates, D., Maechler, M., Bolker, B. and Walker, S. (2015) Fitting linear mixed-effects models using lme4. *J. Stat. Softw.* 67, 1-48.

Burton, E.A. and Walter, L.M. (1987) Relative precipitation rates of aragonite and mg calcite from seawater: temperature or carbonate ion control? *Geology* 15, 111-114. https://doi.org/10.1130/0091-7613(1987)15%3C111:rproaa%3E2.0.co;2

Chamberlain, J.A. (1978) Mechanical-properties of coral skeleton - compressive strength and its adaptive significance. *Paleobiology* 4, 419-435.

Chan, D., Kent, E.C., Berry, D.I. and Huybers, P. (2019) Correcting datasets leads to more homogeneous early-twentieth-century sea surface warming. *Nature* 571, 393-397. https://doi.org/10.1038/s41586-019-1349-2

Chan, N.C.S. and Connolly, S.R. (2013) Sensitivity of coral calcification to ocean acidification: a meta-analysis. *Glob. Change Biol.* 19, 282-290. https://doi.org/10.1111/gcb.12011

Chelton, D.B. and Risien, C.M. (2016) Zonal and Meridional Discontinuities and Other Issues with the HadISST1.1 Dataset (Technical report). https://ir.library.oregonstate.edu/concern/defaults/kw52j9632.

Cohen, A.L. and McConnaughey, T.A. (2003) Geochemical perspectives on coral mineralization. *Rev. Mineral. Geochem.* 54, 151-187. https://doi.org/10.2113/0540151

Cooper, T.F., De 'Ath, G., Fabricius, K.E. and Lough, J.M. (2008) Declining coral calcification in massive Porites in two nearshore regions of the northern Great Barrier Reef. *Glob. Change Biol.* 14, 529-538. https://doi.org/10.1111/j.1365-2486.2007.01520.x

Cooper, T.F., O'Leary, R.A. and Lough, J.M. (2012) Growth of western Australian corals in the Anthropocene. *Science* 335, 593-596. https://doi.org/10.1126/science.1214570

Crook, E.D., Cohen, A.L., Rebolledo-Vieyra, M., Hernandez, L. and Paytan, A. (2013) Reduced calcification and lack of acclimatization by coral colonies growing in areas of persistent natural acidification. *Proc. Nat. Acad. Sci. U.S.A.* 110, 11044-11049. https://doi.org/10.1073/pnas.1301589110

Cyronak, T., Schulz, K.G., Santos, I.R. and Eyre, B.D. (2014) Enhanced acidification of global coral reefs driven by regional biogeochemical feedbacks. *Geophys. Res. Lett.* 41, 5538-5546. https://doi.org/10.1002/2014gl060849

D'Olivo, J.P., McCulloch, M.T., Eggins, S.M. and Trotter, J. (2015) Coral records of reef-water pH across the central Great Barrier Reef, Australia: assessing the influence of river runoff on inshore reefs. *Biogeosciences* 12, 1223-1236. https://doi.org/10.5194/bg-12-1223-2015

D'Olivo, J.P., McCulloch, M.T. and Judd, K. (2013) Long-term records of coral calcification across the central Great Barrier Reef: assessing the impacts of river runoff and climate change. *Coral Reefs* 32, 999-1012. https://doi.org/10.1007/s00338-013-1071-8

De'ath, G., Fabricius, K. and Lough, J. (2013) Yes - Coral calcification rates have decreased in the last twenty-five years! *Mar. Geol.* 346, 400-402. https://doi.org/10.1016/j.margeo.2013.09.008

De'ath, G., Lough, J.M. and Fabricius, K.E. (2009) Declining coral calcification on the Great Barrier Reef. *Science* 323, 116-119. https://doi.org/10.1126/science.1165283

DeCarlo, T.M., Cohen, A.L., Barkley, H.C., Cobban, Q., Young, C., Shamberger, K.E., Brainard, R.E. and Golbuu, Y. (2015) Coral macrobioerosion is accelerated by ocean acidification and nutrients. *Geology* 43, 7-10. https://doi.org/10.1130/g36147.1

Dickson, A.G. (1990) Thermodynamics of the dissociation of boric-acid in synthetic seawater from 273.15 K to 318.15 K. *Deep Sea Res. A* 37, 755-766. https://doi.org/10.1016/0198-0149(90)90004-f

Doney, S.C., Fabry, V.J., Feely, R.A. and Kleypas, J.A. (2009) Ocean acidification: The other CO_2 problem. *Annu. Rev. Mar. Sci.* 1, 169-192. https://doi.org/10.1146/annurev.marine.010908.163834

Dustan, P. (1975) Growth and form in reef-building coral Montastrea annularis. *Mar. Biol.* 33, 101-107. https://doi.org/10.1007/bf00390714

Fantazzini, P., Mengoli, S., Pasquini, L., Bortolotti, V., Brizi, L., Mariani, M., Di Giosia, M., Fermani, S., Capaccioni, B., Caroselli, E., Prada, F., Zaccanti, F., Levy, O., Dubinsky, Z., Kaandorp, J.A., Konglerd, P., Hammel, J.U., Dauphin, Y., Cuif, J.P., Weaver, J.C., Fabricius, K.E., Wagermaier, W., Fratzl, P., Falini, G. and Goffredo, S. (2015) Gains and losses of coral skeletal porosity changes with ocean acidification acclimation. *Nat. Commun.* 6, 7785. https://doi.org/10.1038/ncomms8785

Feely, R.A., Doney, S.C. and Cooley, S.R. (2009) Ocean Acidification: Present conditions and future changes in a high-CO_2 world. *Oceanography* 22, 36-47. https://doi.org/10.5670/oceanog.2009.95

Foster, G.L., Pogge von Strandmann, P.A.E. and Rae, J.W.B. (2010) Boron and magnesium isotopic composition of seawater. *Geochem. Geophys. Geosyst.* 11. https://doi.org/10.1029/2010gc003201

Friedlingstein, P., Jones, M.W., O'Sullivan, M., Andrew, R.M., Hauck, J., Peters, G.P., Peters, W., Pongratz, J., Sitch, S., Le Quere, C., Bakker, D.C.E., Canadell, J.G., Ciais, P., Jackson, R.B., Anthoni, P., Barbero, L., Bastos, A., Bastrikov, V., Becker, M., Bopp, L., Buitenhuis, E., Chandra, N., Chevallier, F., Chini, L.P., Currie, K.I., Feely, R.A., Gehlen, M., Gilfillan, D., Gkritzalis, T., Goll, D.S., Gruber, N., Gutekunst, S., Harris, I., Haverd, V., Houghton, R.A., Hurtt, G., Ilyina, T., Jain, A.K., Joetzjer, E., Kaplan, J.O., Kato, E., Goldewijk, K.K., Korsbakken, J.I., Landschutzer, P., Lauvset, S.K., Lefevre, N., Lenton, A., Lienert, S., Lombardozzi, D., Marland, G., McGuire, P.C., Melton, J.R., Metzl, N., Munro, D.R., Nabel, J., Nakaoka, S.I., Neill, C., Omar, A.M., Ono, T., Peregon, A., Pierrot, D., Poulter, B., Rehder, G., Resplandy, L., Robertson, E., Rodenbeck, C., Seferian, R., Schwinger, J., Smith, N., Tans, P.P., Tian, H.Q., Tilbrook, B., Tubiello, F.N., van der Werf, G.R., Wiltshire, A.J. and Zaehle, S. (2019) Global Carbon Budget 2019. *Earth System Science Data* 11, 1783-1838. https://doi.org/10.5194/essd-11-1783-2019

Gagliano, M., McCormick, M.I., Moore, J.A. and Depczynski, M. (2010) The basics of acidification: baseline variability of pH on Australian coral reefs. *Mar. Biol.* 157, 1849-1856. https://doi.org/10.1007/s00227-010-1456-y

Guo, W. (2019) Seawater temperature and buffering capacity modulate coral calcifying pH. *Sci. Rep.* 9, 1189. https://doi.org/10.1038/s41598-018-36817-y

Hoegh-Guldberg, O., Mumby, P.J., Hooten, A.J., Steneck, R.S., Greenfield, P., Gomez, E., Harvell, C.D., Sale, P.F., Edwards, A.J., Caldeira, K., Knowlton, N., Eakin, C.M., Iglesias-Prieto, R., Muthiga, N., Bradbury, R.H., Dubi, A. and Hatziolos, M.E. (2007) Coral reefs under rapid climate change and ocean acidification. *Science* 318, 1737-1742. https://doi.org/10.1126/science.1152509

Hurrell, J.W., Holland, M.M., Gent, P.R., Ghan, S., Kay, J.E., Kushner, P.J., Lamarque, J.F., Large, W.G., Lawrence, D., Lindsay, K., Lipscomb, W.H., Long, M.C., Mahowald, N., Marsh, D.R., Neale, R.B., Rasch, P., Vavrus, S., Vertenstein, M., Bader, D., Collins, W.D., Hack, J.J., Kiehl, J. and Marshall, S. (2013) The Community Earth System Model A Framework for Collaborative Research. *Bull. Am. Meteorol. Soc.* 94, 1339-1360. https://doi.org/10.1175/bams-d-12-00121.1

Huston, M. (1985) Variation in coral growth-rates with depth at Discovery Bay, Jamaica. *Coral Reefs* 4, 19-25. https://doi.org/10.1007/bf00302200

Klochko, K., Kaufman, A.J., Yao, W.S., Byrne, R.H. and Tossell, J.A. (2006) Experimental measurement of boron isotope fractionation in seawater. *Earth Planet. Sci. Lett.* 248, 276-285. https://doi.org/10.1016/j.epsl.2006.05.034

Liu, Y., Peng, Z.C., Zhou, R.J., Song, S.H., Liu, W.G., You, C.F., Lin, Y.P., Yu, K.F., Wu, C.C., Wei, G.J., Xie, L.H., Burr, G.S. and Shen, C.C. (2014) Acceleration of modern acidification in the South China Sea driven by anthropogenic CO_2. *Sci. Rep.* 4, 5148. https://doi.org/10.1038/srep05148

Lough, J.M. and Barnes, D.J. (2000) Environmental controls on growth of the massive coral *Porites*. *J. Exp. Mar. Biol. Ecol.* 245, 225-243. https://doi.org/10.1016/s0022-0981(99)00168-9

Lough, J.M. and Cantin, N.E. (2014) Perspectives on massive coral growth rates in a changing ocean. *Biol. Bull.* 226, 187-202. https://doi.org/10.1086/BBLv226n3p187

Lough, J.M. and Cooper, T.F. (2011) New insights from coral growth band studies in an era of rapid environmental change. *Earth Sci. Rev.* 108, 170-184. https://doi.org/10.1016/j.earscirev.2011.07.001

Madin, J.S. (2004) A mechanistic approach to understanding and predicting hydrodynamic disturbance on coral reefs., Department of Marine Biology and Aquaculture. James Cook University.

Madin, J.S., Hughes, T.P. and Connolly, S.R. (2012) Calcification, storm damage and population resilience of tabular corals under climate change. *Plos One* 7, e46637. https://doi.org/10.1371/journal.pone.0046637

Madin, J.S., O'Donnell, M.J. and Connolly, S.R. (2008) Climate-mediated mechanical changes to post-disturbance coral assemblages. *Biology Letters* 4, 490-493. https://doi.org/10.1098/rsbl.2008.0249

Martinez, A., Crook, E.D., Barshis, D.J., Potts, D.C., Rebolledo-Vieyra, M., Hernandez, L. and Paytan, A. (2019) Species-specific calcification response of Caribbean corals after 2-year transplantation to a low aragonite saturation submarine spring. *Proceedings of the Royal Society B-Biological Sciences* 286. https://doi.org/10.1098/rspb.2019.0572

McCulloch, M., Falter, J., Trotter, J. and Montagna, P. (2012) Coral resilience to ocean acidification and global warming through pH up-regulation. *Nat. Clim. Change* 2, 623-633. https://doi.org/10.1038/nclimate1473

McCulloch, M.T., D'Olivo, J.P., Falter, J., Holcomb, M. and Trotter, J.A. (2017) Coral calcification in a changing World and the interactive dynamics of pH and DIC upregulation. *Nat. Commun.* 8, 15686. https://doi.org/10.1038/ncomms15686

Mollica, N.R., Guo, W.F., Cohen, A.L., Huang, K.F., Foster, G.L., Donald, H.K. and Solow, A.R. (2018) Ocean acidification affects coral growth by reducing skeletal density. *Proc. Nat. Acad. Sci. U.S.A.* 115, 1754-1759. https://doi.org/10.1073/pnas.1712806115

Mongin, M., Baird, M.E., Tilbrook, B., Matear, R.J., Lenton, A., Herzfeld, M., Wild-Allen, K., Skerratt, J., Margvelashvili, N., Robson, B.J., Duarte, C.M., Gustafsson, M.S.M., Ralph, P.J. and Steven, A.D.L. (2016) The exposure of the Great Barrier Reef to ocean acidification. *Nat. Commun.* 7, 10732. https://doi.org/10.1038/ncomms10732

Nothdurft, L.D. and Webb, G.E. (2007) Microstructure of common reef-building coral genera *Acropora*, *Pocillopora*, *Goniastrea* and *Porites*: constraints on spatial resolution in geochemical sampling. *Facies* 53, 1-26. https://doi.org/10.1007/s10347-006-0090-0

Orr, J.C., Fabry, V.J., Aumont, O., Bopp, L., Doney, S.C., Feely, R.A., Gnanadesikan, A., Gruber, N., Ishida, A., Joos, F., Key, R.M., Lindsay, K., Maier-Reimer, E., Matear, R., Monfray, P., Mouchet, A., Najjar, R.G., Plattner, G.K., Rodgers, K.B., Sabine, C.L., Sarmiento, J.L., Schlitzer, R., Slater, R.D., Totterdell, I.J., Weirig, M.F., Yamanaka, Y. and Yool, A. (2005) Anthropogenic ocean acidification over the twenty-first century and its impact on calcifying organisms. *Nature* 437, 681-686. https://doi.org/10.1038/nature04095

Pandolfi, J.M. (2015) Incorporating uncertainty in predicting the future response of coral reefs to climate change. *Annu. Rev. Ecol. Evol. Syst.* 46, 281-303. https://doi.org/10.1146/annurev-ecolsys-120213-091811

Pandolfi, J.M., Connolly, S.R., Marshall, D.J. and Cohen, A.L. (2011) Projecting coral reef futures under global warming and ocean acidification. *Science* 333, 418-422. https://doi.org/10.1126/science.1204794

Pelejero, C., Calvo, E., McCulloch, M.T., Marshall, J.F., Gagan, M.K., Lough, J.M. and Opdyke, B.N. (2005) Preindustrial to modern interdecadal variability in coral reef pH. *Science* 309, 2204-2207. https://doi.org/10.1126/science.1113692

Rayner, N.A., Parker, D.E., Horton, E.B., Folland, C.K., Alexander, L.V., Rowell, D.P., Kent, E.C. and Kaplan, A. (2003) Global analyses of sea surface temperature, sea ice, and night marine air temperature since the late nineteenth century. *J. Geophys. Res.-Atmos.* 108. https://doi.org/10.1029/2002jd002670

Ridd, P.V., da Silva, E.T. and Stieglitz, T. (2013) Have coral calcification rates slowed in the last twenty years? *Mar. Geol.* 346, 392-399. https://doi.org/10.1016/j.margeo.2013.09.002

Rippe, J.P., Baumann, J.H., De Leener, D.N., Aichelman, H.E., Friedlander, E.B., Davies, S.W. and Castillo, K.D. (2018) Corals sustain growth but not skeletal density across the Florida Keys Reef Tract despite ongoing warming. *Glob. Change Biol.* 24, 5205-5217. https://doi.org/10.1111/gcb.14422

Scott, P.J.B. and Risk, M.J. (1988) The effect of *lithophaga* (bivalvia, mytilidae) boreholes on the strength of the coral *Porites lobata*. *Coral Reefs* 7, 145-151. https://doi.org/10.1007/bf00300974

Su, R.X., Lough, J.M. and Sun, D.H. (2016) Variations in massive *Porites* growth rates at Hainan Island, northern South China Sea. *Mar. Ecol. Prog. Ser.* 546, 47-60. https://doi.org/10.3354/meps11654

Tambutte, E., Venn, A.A., Holcomb, M., Segonds, N., Techer, N., Zoccola, D., Allemand, D. and Tambutte, S. (2015) Morphological plasticity of the coral skeleton under CO_2-driven seawater acidification. *Nat. Commun.* 6, 7368. https://doi.org/10.1038/ncomms8368

Taylor, R.B., Barnes, D.J. and Lough, J.M. (1993) Simple-models of density band formation in massive corals. *J. Exp. Mar. Biol. Ecol.* 167, 109-125. https://doi.org/10.1016/0022-0981(93)90187-s

Tomascik, T. and Sander, F. (1985) Effects of eutrophication on reef-building corals .I. Growth rate of the reef-building coral Montastrea annularis. *Mar. Biol.* 87, 143-155. https://doi.org/10.1007/bf00539422

Trotter, J., Montagna, P., McCulloch, M., Silenzi, S., Reynaud, S., Mortimer, G., Martin, S., Ferrier-Pagès, C., Gattuso, J.-P. and Rodolfo-Metalpa, R. (2011) Quantifying the pH 'vital effect' in the temperate zooxanthellate coral *Cladocora caespitosa*: Validation of the boron seawater pH proxy. *Earth Planet. Sci. Lett.* 303, 163-173. https://doi.org/10.1016/j.epsl.2011.01.030

Uthicke, S., Furnas, M. and Lonborg, C. (2014) Coral reefs on the edge? Carbon chemistry on inshore reefs of the Great Barrier Reef. *Plos One* 9, e109092. https://doi.org/10.1371/journal.pone.0109092

Wei, G.J., McCulloch, M.T., Mortimer, G., Deng, W.F. and Xie, L.H. (2009) Evidence for ocean acidification in the Great Barrier Reef of Australia. *Geochim. Cosmochim. Acta* 73, 2332-2346. https://doi.org/10.1016/j.gca.2009.02.009

Wei, G.J., Wang, Z.B., Ke, T., Liu, Y., Deng, W.F., Chen, X.F., Xu, J.F., Zeng, T. and Xie, L.H. (2015) Decadal variability in seawater pH in the West Pacific: Evidence from coral records. *J. Geophys. Res.-Oceans* 120, 7166-7181. https://doi.org/10.1002/2015jc011066

Wisshak, M., Schonberg, C.H.L., Form, A. and Freiwald, A. (2012) Ocean acidification accelerates reef bioerosion. *Plos One* 7. https://doi.org/10.1371/journal.pone.0045124

Wood, S.N. (2011) Fast stable restricted maximum likelihood and marginal likelihood estimation of semiparametric generalized linear models. *J. R. Stat. Soc. Ser. B* 73, 3-36.

Wood, S.N. (2017) Generalized Additive Models: An Introduction with R (2nd edition). Chapman and Hall/CRC.

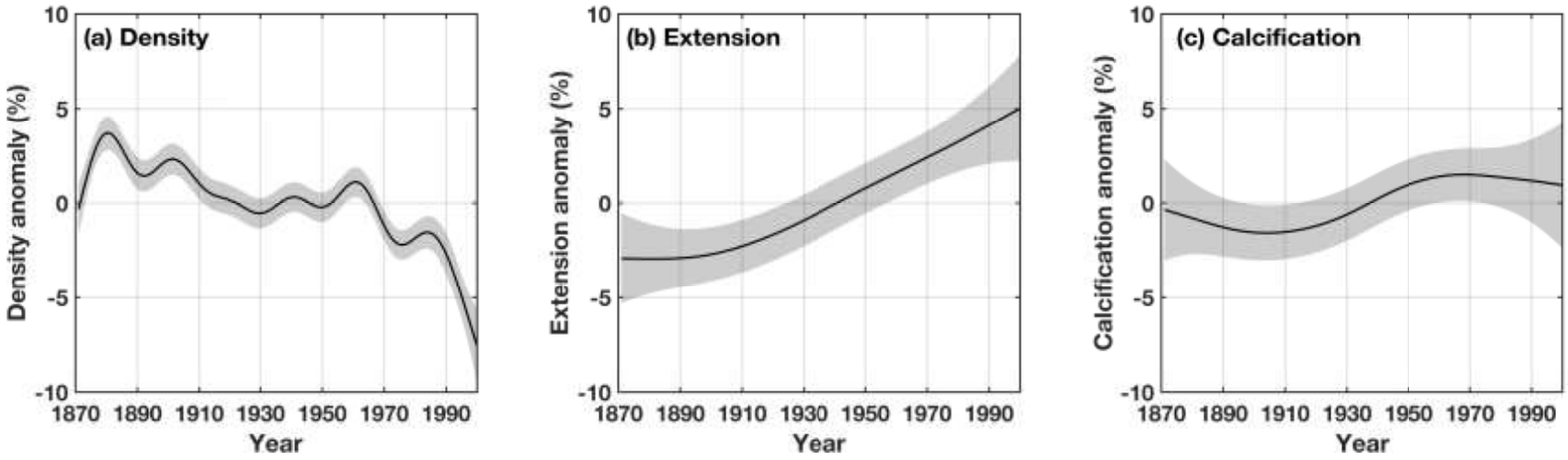

Fig. 1. Partial-effects plots showing changes in *Porites* skeletal parameters at the Great Barrier Reef from 1871 to 2000: **(a)** density, **(b)** extension, and **(c)** calcification. The temporal trends were derived from 60 long (≥50 years) *Porites* skeletal cores from 39 reefs across the Great Barrier Reef (Fig. S1, Methods, Text S3, De'ath et al., 2009), and are expressed as the anomalies relative to the corresponding mean values over 1951-1960, a common period that all cores cover. The gray bands indicate 95% confidence intervals for the predicted value for any given year.

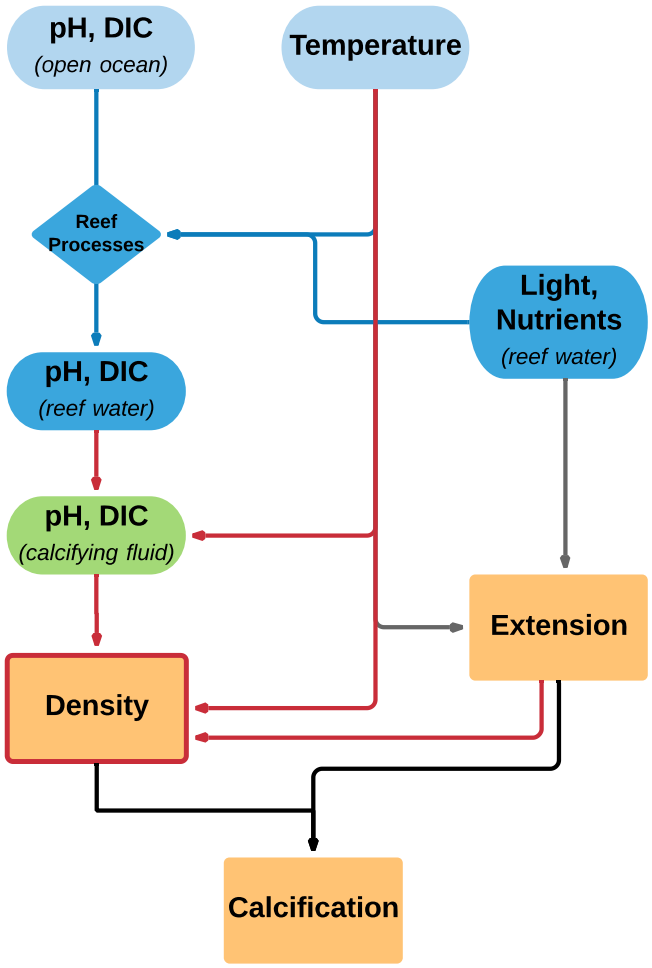

Fig. 2. Relations between open ocean and reef water carbonate chemistry and their influences on *Porites* coral skeletal growth. *Porites* skeletal density is directly affected by the carbonate chemistry of its calcifying fluid, which in turn is influenced by reef-seawater chemistry (Mollica et al., 2018; Guo, 2019, Text S1). This makes *Porites* skeletal density most sensitive to ocean acidification. Besides seawater chemistry, *Porites* skeletal density is also directly affected by seawater temperature and the rate of skeletal extension (Mollica et al., 2018), while other environmental factors (e.g., light condition, nutrient levels) affect *Porites* skeletal density indirectly through their influences on the skeletal extension and/or reef-water chemistry. We use a coral skeletal growth model to isolate the effects of different factors and quantify their respective contributions to the coral growth (Equation 1, Fig. 3). Arrows denote the processes that are explicitly (red) or implicitly (gray) simulated in our coral skeletal growth model, and the factors influencing reef water carbonate chemistry (blue).

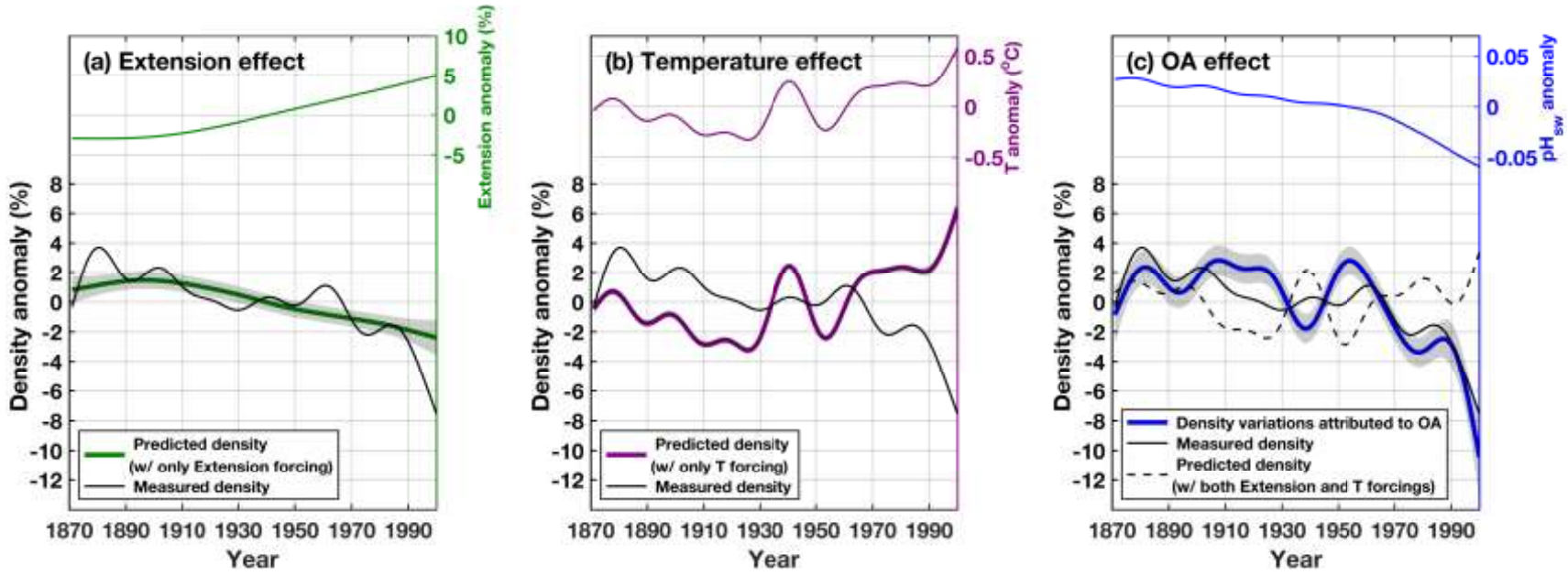

Fig. 3. Contributions of different factors to the changes in *Porites* skeletal density at the Great Barrier Reef: **(a)** extension, **(b)** temperature, and **(c)** ocean acidification. The impact of ocean acidification is determined by subtracting the model predicted effects of extension and temperature (dashed line, panel c) from the measured density (black line, Equation 1). Also shown for comparison are the temporal changes in each factor (upper panels). All anomalies are calculated relative to the corresponding mean values over 1951-1960, a common period that all cores cover, and the seawater temperature and pH are derived from the HadiSST dataset and the CESM-BGC historical run, respectively (Methods). The model-predicted high-frequency density variability around 1940 (b-c) likely arises from the uncertainties in the historical temperature data-products during this time period (see text for details).

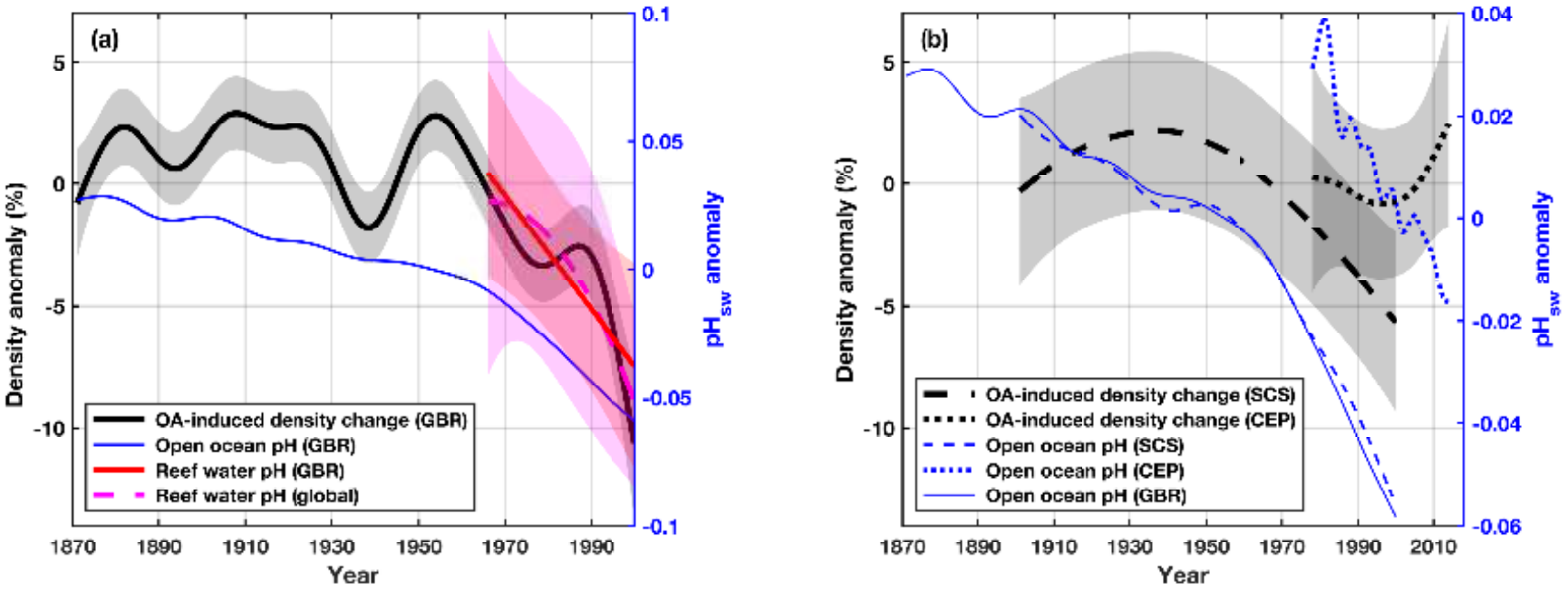

Fig. 4. Impacts of OA on *Porites* skeletal density at different reef systems and their correlations with ocean and reef-water pH: **(a)** Great Barrier Reef (GBR), **(b)** South China Sea (SCS) and central equatorial Pacific (CEP) reefs. The declines in reef-water pH at the GBR (Methods, Pelejero et al., 2005; Wei et al., 2009; D'Olivo et al., 2015) and other reefs around the globe (Cyronak et al., 2014) are shown in (a). Also shown for comparison are the open-ocean pH derived from the Community Earth System Model Biogeochemical (CESM-BGC) historical run. The gray and colored bands indicate 95% confidence intervals.