

# The synergistic effects of ocean acidification and organic metabolism on calcium carbonate ( $\text{CaCO}_3$ ) dissolution in coral reef sediments



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## ABSTRACT

Ocean acidification (OA) is expected to reduce the net ecosystem calcification (NEC) rates and overall accretion of coral reef ecosystems. However, despite the fact that sediments are the most abundant form of calcium carbonate ( $\text{CaCO}_3$ ) in coral reef ecosystems and their dissolution may be more sensitive to OA than biogenic calcification, the impacts of OA induced sediment dissolution on coral reef NEC rates and  $\text{CaCO}_3$  accretion are poorly constrained. Carbon dioxide addition and light attenuation experiments were performed at Heron Island, Australia in an attempt to tease apart the influence of OA and organic metabolism (e.g. respiratory  $\text{CO}_2$  production) on  $\text{CaCO}_3$  dissolution. Overall,  $\text{CaCO}_3$  dissolution rates were an order of magnitude more sensitive to elevated  $\text{CO}_2$  and decreasing seawater aragonite saturation state ( $\Omega_{\text{Ar}}$ ; 300–420% increase in dissolution per unit decrease in  $\Omega_{\text{Ar}}$ ) than published reductions in biologically mediated calcification due to OA. Light attenuation experiments led to a 70% reduction in net primary production (NPP), which subsequently induced an increase in daytime (~115%) and net diel (~375%)  $\text{CaCO}_3$  dissolution rates. High  $\text{CO}_2$  and low light acted in synergy to drive a ~575% increase in net diel dissolution rates. Importantly, disruptions to the balance of photosynthesis and respiration (P/R) had a significant effect on daytime  $\text{CaCO}_3$  dissolution, while average water column  $\Omega_{\text{Ar}}$  was the main driver of nighttime dissolution rates. A simple model of platform-integrated dissolution rates was developed demonstrating that seasonal changes in photosynthetically active radiation (PAR) can have an important effect on platform integrated  $\text{CaCO}_3$  sediment dissolution rates. The considerable response of  $\text{CaCO}_3$  sediment dissolution to elevated  $\text{CO}_2$  means that much of the response of coral reef communities and ecosystems to OA could be due to increases in  $\text{CaCO}_3$  sediment and framework dissolution, and not decreases in biogenic calcification.

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## 1. Introduction

Ocean acidification (OA), or the ‘other  $\text{CO}_2$  problem’, refers to the rising seawater  $p\text{CO}_2$  and decreasing pH of oceanic ecosystems (Caldeira and Wickett, 2003; Doney et al., 2009). The chemical effects of increasing  $\text{CO}_2$  in seawater are well described and result in a decrease in seawater pH and carbonate ( $\text{CO}_3^{2-}$ ) ion concentration (Zeebe and Wolf-Gladrow, 2001). Due to a reduction in  $\text{CO}_3^{2-}$  concentration, OA also lowers the calcium carbonate ( $\text{CaCO}_3$ ) saturation state ( $\Omega$ ) of seawater (Zeebe and Wolf-Gladrow, 2001). When seawater  $\Omega > 1$  precipitation of  $\text{CaCO}_3$  is thermodynamically favorable, while at  $\Omega < 1$  dissolution becomes thermodynamically favored (Morse and Mackenzie, 1990). Due to a reduction in both pH and  $\Omega$ , it is expected that OA will have negative effects on coral reef  $\text{CaCO}_3$  production, accumulation, and loss processes (Hoegh-Guldberg et al., 2007; Chan and Connolly, 2012; Cyronak et al., 2015). Most current predictions of how OA will affect coral reef  $\text{CaCO}_3$  budgets have focused on the relationship between water column  $\Omega$

and calcification rates (i.e. biogenic  $\text{CaCO}_3$  production) of corals or coral reef communities (Gattuso et al., 1998a; Shamberger et al., 2011). However, net ecosystem calcification (NEC) of coral reefs is the net result of both  $\text{CaCO}_3$  production (e.g. calcification) and loss (e.g.  $\text{CaCO}_3$  dissolution) processes (Andersson and Gledhill, 2013; Eyre et al., 2014). Therefore, to understand how OA will affect coral reef NEC and accretion it becomes important to determine how changes in seawater  $\Omega$  affects both  $\text{CaCO}_3$  formation and loss processes, which most likely respond differently to OA (Andersson et al., 2009).

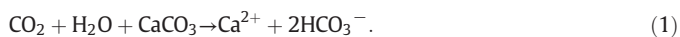
$\text{CaCO}_3$  found in coral reefs can be thought of as existing in two main pools; (1)  $\text{CaCO}_3$  framework (e.g. intact coral skeletons which may or may not be covered by living coral and other organisms) and (2) permeable  $\text{CaCO}_3$  sediments (e.g. broken down  $\text{CaCO}_3$  formed from reef framework and other sources of  $\text{CaCO}_3$  such as calcifying algae and infaunal organisms). Permeable sediments represent the buildup of  $\text{CaCO}_3$  over thousands of years and can make up the majority of  $\text{CaCO}_3$  stored in modern coral reef ecosystems (Gattuso et al., 1998b; Smith et al., 2009). In fact, some modern reef structures such as shallow reef flats, lagoons, and coral cays were largely formed due to a stagnation in sea level rise over the past ~6000 years and constant supply of  $\text{CaCO}_3$  sediment from the breakdown of reef framework (Gourlay,

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1988; Ryan et al., 2001). Therefore, any increase in the dissolution of CaCO<sub>3</sub> sediments due to OA will not only have a large impact on NEC rates, but also affect the formation of valuable habitat both above and below the high tide mark. As such, understanding the dissolution of CaCO<sub>3</sub> sediments in an acidifying ocean could be as, or more important than understanding changes to biogenic calcification when examining the effects of OA on coral reef ecosystems (Eyre et al., 2014).

Dissolution of CaCO<sub>3</sub> in permeable sediments is largely a function of porewater chemistry, which is controlled by the carbonate chemistry of the overlying seawater and any biogeochemical reactions occurring within the sediments (Ku et al., 1999; Burdige and Zimmerman, 2002; Cyronak et al., 2013a). In permeable sediments such as the calcium carbonate sands found on coral reefs, advection of overlying seawater into and out of the sediments plays an important role in regulating benthic fluxes and metabolism (Rao et al., 2012; Cyronak et al., 2013a; Huettel et al., 2014). Advection increases the transport of overlying seawater into the sediments and the export of porewater out, increasing solute exchange above that of diffusive conditions (Precht and Huettel, 2003). Therefore, advection can have a dramatic influence on porewater carbonate chemistry by enhancing the input of surface waters into the sediments (Cyronak et al., 2013a; Drupp et al., 2016) as well as by influencing biological activity via the input of organic and inorganic matter (Wild et al., 2004b; Cook and Røy, 2006). A reduction of  $\Omega$  in seawater overlying permeable sediments (i.e. ocean acidification) results in a starting point closer to the solubility of CaCO<sub>3</sub> minerals when that seawater is flushed through the sediments via advective processes (termed environmental dissolution) (Eyre et al., 2014). For example, it has been shown that CO<sub>2</sub> concentrations predicted by the end of the century for the open ocean could increase the net dissolution of permeable coral reef sediments ~4 times above current rates, and that porewater advection enhances the effects of OA (Cyronak et al., 2013a). The addition of CO<sub>2</sub> to the porewaters through oxidic respiration can drive further CaCO<sub>3</sub> dissolution (termed metabolic dissolution) in the following net reaction (Burdige and Zimmerman, 2002):



Therefore, it is important to determine how both OA and benthic metabolic processes act in synergy to drive CaCO<sub>3</sub> dissolution in permeable coral reef sediments.

Net ecosystem calcification of coral reefs follows a diel trend increasing with light availability, most likely due to the strong influence of net ecosystem production (NEP) on NEC (Shaw et al., 2012; Koweek et al., 2014; Silverman et al., 2014). The correlation between NEC and NEP may be related to the phenomenon of light enhanced calcification, which has been well documented in corals (Goreau, 1959; Moya et al., 2006; Allemand et al., 2011). However, much less is known about the effects of light availability and changes in organic metabolism on CaCO<sub>3</sub> precipitation and dissolution in permeable coral reef sediments. There are diel trends in CaCO<sub>3</sub> precipitation/dissolution in CaCO<sub>3</sub> sediments (Yates and Halley, 2006; Cyronak et al., 2013b), which are most likely due changes in porewater chemistry driven by photosynthesis and respiration (Schoon et al., 2010). Under ambient pCO<sub>2</sub> conditions a strong correlation exists between sediment net primary production (NPP) and CaCO<sub>3</sub> precipitation/dissolution (Rao et al., 2012; Cyronak et al., 2013b). However, little is known about the combined effects of increased water column pCO<sub>2</sub> (OA) and changes in organic metabolism (i.e. photosynthesis and respiration). A simple model calculation suggests that dissolution will increase with depth due to the effects of light attenuation on NPP (Cyronak et al., 2013a). However, to our knowledge, no experimental study has combined OA conditions and manipulations of NPP in situ in permeable coral reef sands.

We hypothesize that reduced light will result in a reduction in sediment NPP and increased CaCO<sub>3</sub> dissolution due to a decrease in porewater  $\Omega$ . In addition, the synergistic effects of OA and reduced light will result in enhanced environmental and metabolic dissolution

due to further reductions in porewater  $\Omega$ . We attempt to tease apart the influence of CO<sub>2</sub> additions to the water column and changes in sediment NPP via light attenuation on the dissolution of CaCO<sub>3</sub> in coral reef sediments using in situ advective benthic chambers at Heron Island, Great Barrier Reef (GBR). This will add insight to our understanding of how CaCO<sub>3</sub> dissolution changes under natural (e.g. seasonal and depth effects) and anthropogenically (e.g. sediment inputs and pelagic algal growth) induced variations in light availability, as well as any other perturbations that may alter benthic organic metabolism, in combination with OA.

## 2. Materials and methods

### 2.1. Study site

Heron Island is a coral cay in the Capricorn and Bunker Group of coral reefs situated at the southern end of the Great Barrier Reef (GBR). The island is comprised mainly of CaCO<sub>3</sub> sands and is relatively small (~0.16 km<sup>2</sup>) compared to the surrounding lagoon and reef flat (~26.4 km<sup>2</sup>). The reef flat and lagoon are dominated by sandy habitats, which make up ~75% of benthic areal coverage throughout the Heron Island platform (Wild et al., 2004a). Our study site was the same as in many previous studies (Eyre et al., 2008, 2013; Glud et al., 2008; Cyronak et al., 2013a, 2013b), and is a well characterized sandy patch located within the reef flat along the southeastern end of the island. The sediment has a median grain size of 0.83 mm, porosity of 0.52–0.57 (vol:vol), and is highly permeable ( $1.6\text{--}6.0 \times 10^{-11} \text{ m}^{-2}$ ) (Glud et al., 2008; Cyronak et al., 2013b). The major CaCO<sub>3</sub> minerals present in the sediment are aragonite (65%) and high magnesium calcite (HMC; 33%) with a mole % MgCO<sub>3</sub> of 15.2%, which is typical for tropical reef systems (Weber and Woodhead, 1969; Cyronak et al., 2013b). Over the course of this study the water column depth at the study site ranged from 0.2–2.2 m, with an average depth of 0.9 m.

### 2.2. Chamber incubations

A series of incubations were conducted over 3 consecutive days (19–21 May 2014), each of which started at dawn and lasted for ~23 h in order to cover a full diel cycle. Advective benthic chambers identical to the ones in Cyronak et al. (2013a) were used to determine rates of photosynthesis/respiration and CaCO<sub>3</sub> precipitation/dissolution in the sediments. The plexiglass chambers have an internal diameter of 19 cm and cover 0.028 m<sup>2</sup> of sediment, enclosing ~4–5 L of seawater to a height of 15–20 cm above the sediment surface during each incubation. The chamber bases were inserted ~15 cm into sediment free of any visible macrophytes or macrofauna and allowed to equilibrate for ~1 h before beginning any incubations. A horizontal spinning disk within each chamber induces advective flow similar to in situ, flow- and topography-induced advection (Huettel et al., 1996). During each incubation the disk was set to spin at 40 rotations per minute (RPM), which induces an advective exchange rate of ~43 L m<sup>-2</sup> day<sup>-1</sup> between the sediment and water column at this site (Glud et al., 2008).

In order to tease apart the influence of organic metabolism and ocean acidification, three in situ manipulative treatments were conducted; high CO<sub>2</sub> (+CO<sub>2</sub>), low light (LL), and low light plus high CO<sub>2</sub> (LL + CO<sub>2</sub>). Low light treatments were achieved by covering the chambers in shade cloth, which resulted in a ~75% reduction from ambient levels of integrated daytime photosynthetically active radiation (PAR) (Fig. 1). High CO<sub>2</sub> treatments were conducted according to the methods of Cyronak et al. (2013a). Briefly, a silicone tube inside each +CO<sub>2</sub> and LL + CO<sub>2</sub> treatment chamber was pressurized with 99.9% CO<sub>2</sub> gas until the desired pH within the chamber (~0.2 pH units lower than control) was reached. Duplicates of the three treatments plus control chambers were run each day resulting in a total of 8 incubations every day. At the end of each incubation the chambers were moved ~1 m to undisturbed sediment prior to starting the next incubation. The 3 days

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