



RESEARCH LETTER

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Key Points:

- We analyzed carbonate chemistry measurements, recorded weekly between 2008 and 2014 in the Chuuk lagoon in the western Pacific Ocean
- Under the longer water residence time conditions, calcification and respiration together lowered the pH of the reef water
- A strong link was found between reef water carbonate chemistry and large-scale climatic variations

Supporting Information:

- Supporting Information S1
- Dataset S1

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Influence of ambient water intrusion on coral reef acidification in the Chuuk lagoon, located in the coral-rich western Pacific Ocean

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Abstract Weekly carbonate chemistry condition data recorded between 2008 and 2014 in the Chuuk lagoon (7.3°N and 151.5°E) of the Federated States of Micronesia, located in the western Pacific Ocean, were analyzed. The results showed that, during periods of weak intrusion of ambient seawater from the surrounding open ocean, two internal biological processes (calcification and respiration) reinforced each other and together lowered the pH of the reef water for extended periods, ranging from a few to several months. The analysis indicated that reduced intrusion of ambient water is associated with periods of low wind speeds. Such conditions increase the residence time of reef water, thus promoting acidification by respiration and calcification. This phenomenon likely affects many other areas of the coral-rich western Pacific Ocean, which contains 50% of global coral reefs and in which the degree of ambient water intrusion into the atolls has been shown to be closely associated with the El Niño–Southern Oscillation-induced wind speed change.

1. Introduction

Coral reefs are a key marine ecosystem, contributing one quarter of the accumulated CaCO₃ in the global ocean [Milliman, 1993; Iglesias-Rodriguez et al., 2002]. They play multiple roles, providing shelters for diverse marine species and providing fisheries and tourism for humans [Moberg and Folke, 1999]. Although highly valued, uniquely positioned in the global marine ecosystem, and persisting over long geological periods, coral reefs are particularly vulnerable to anthropogenic acidification, driven mainly by the oceanic uptake of anthropogenic CO₂ [Hoegh-Guldberg et al., 2007; Andersson and Gledhill, 2013; Albright et al., 2016]. The oceanic uptake of CO₂ decreases the pH of seawater and the degree of saturation of seawater with respect to aragonite or calcitic CaCO₃ (Ω_{AR} or $\Omega_{CAL} = [Ca^{2+}][CO_3^{2-}]/K_A^*_{SP}$, where $K_A^*_{SP}$ is the stoichiometric solubility product for aragonite or calcite). The decrease in aragonite saturation state (Ω_{AR}) is correlated with the decrease in calcification rate of corals [Kleypas et al., 1999; Langdon and Atkinson, 2005; Hoegh-Guldberg et al., 2007] and an increase in coral erosion [Silbiger et al., 2014]. As more anthropogenic CO₂ enters the ocean, the progressive decrease in Ω_{AR} is expected to have adverse effects on coral reefs.

Research over the past two decades has shown that large regional fluctuations in climatic variables (e.g., wind speed, temperature, and precipitation) associated with the El Niño–Southern Oscillation (ENSO) have been superimposed on the decrease in ocean carbonate chemistry (e.g., pH and Ω_{AR}). This is the case in the western tropical Pacific Ocean, which includes half of the global extent of coral reefs, and thus makes it the largest coral habitat worldwide [Spalding et al., 2001]. As recorded in coral skeletal $\delta^{18}O$ records from western Pacific Ocean corals (for example), large changes in sea surface temperature (SST) and salinity occur during ENSO events [McGregor and Gagan, 2004; Suzuki et al., 2007]. Anomalous wind patterns during ENSO events are often related to these changes, but changes in wind speed can also alter the exchange between open ocean and reef waters, and hence the residence time of water within a reef environment; the influence of such wind speed changes is likely to be more pronounced in reef systems having limited exchange of ambient water with the surrounding open ocean [Delesalle and Sournia, 1992; Lowe et al., 2009]. The effect of ambient water intrusion associated with ENSO-induced wind speed variations on reef water biogeochemistry can best be studied in the western tropical Pacific Ocean, where this effect has been shown to be large.

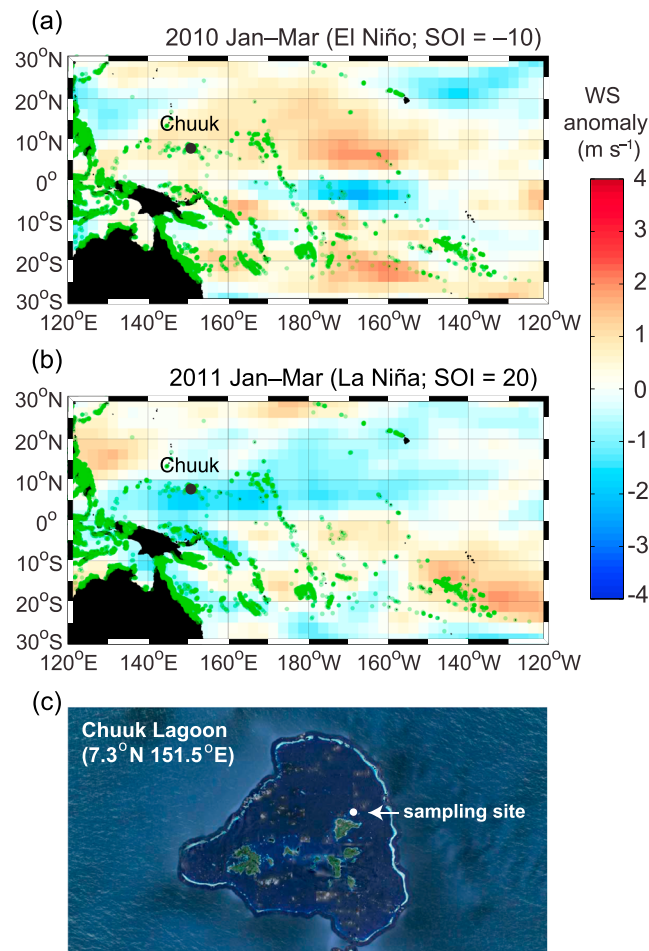


Figure 1. Wind speed anomalies (m s^{-1}) (means of January–March for 2010 and 2011 minus the 6 year mean of the same months) during (a) El Niño (January–March 2010) and (b) La Niña (January–March 2011) periods from the 6 year means of the corresponding months. Positive and negative anomalies are indicated in red and blue, respectively. Gridded wind speed data were obtained from National Centers for Environmental Prediction (NCEP) Reanalysis data (<http://www.esrl.noaa.gov/psd/>). Green dots indicate locations of coral reefs obtained from United Nations Environment Programme World Conservation Monitoring Centre (available at <http://data.unep-wcmc.org/datasets/1>). (c) A satellite image showing the Chuuk lagoon (source: Google Earth[®]) and the sampling site (white circle).

chemistry of such systems. Data from this type of observational study have been reported for the Bermuda coral reef system of the North Atlantic Ocean, where acidification events were driven by offshore productivity changes associated with the negative winter North Atlantic Oscillation index [Yeakel *et al.*, 2015].

The primary goal of the present study was to explore the effects of large-scale climate variations as well as the physical and biological processes on the carbonate chemistry of reef waters. This investigation is based on 6 years of weekly observations of parameters associated with the carbonate system in the Chuuk coral lagoon, located in the coral-rich tropical western Pacific Ocean.

2. Methods

2.1. The Chuuk Lagoon, Western Pacific Ocean

The Chuuk lagoon is one of the largest semiclosed atolls in the western Pacific Ocean (7.3°N and 151.5°E). The lagoon has an area of 2131.5 km^2 , a latitudinal length of 50 km, and a maximum depth of 91 m (Figure 1). The

However, except for one rare report, that of Pelejero *et al.* [2005], we are unaware of any observational evidence that shows a link between reef water carbonate chemistry and ENSO-induced wind speed variations in the coral-rich, western tropical Pacific Ocean. In the above study, interdecadal variability in pH (derived from coral core ^{11}B data) in the semiclosed Flinders Reef (near the Great Barrier Reef) was shown to be influenced by changes in the residence time of its reef water. This occurred because of changes in the strength of the Pacific trade wind, which are primarily associated with variations in the Interdecadal Pacific Oscillation (IPO) index [Pelejero *et al.*, 2005]. However, pH data alone are insufficient to accurately identify the drivers of changes in pH; an additional carbonate parameter is needed. Moreover, data with a coarse time resolution cannot be used to examine seasonal to interannual changes in reef water chemistry associated with ENSO-induced wind speed changes.

Therefore, long-term observations recorded with a finer time resolution (monthly or shorter), of carbonate parameters (i.e., total alkalinity, A_T = the excess of bases formed from weak acids having $K \leq 10^{-4.5}$ over acids having $K > 10^{-4.5}$; and total dissolved inorganic carbon, $C_T = [\text{CO}_2] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}]$) are needed for coral reef systems, in conjunction with basin-scale climate variations (i.e., ENSO, IPO, and North Atlantic Oscillation), in order to estimate the contribution of natural climate variations to changes in the carbonate

SST in the lagoon remains $>28^{\circ}\text{C}$ throughout the year; hence, the lagoon is included in the western Pacific warm pool [Yan *et al.*, 1992]. The eastward flowing North Equatorial Counter Current greatly influences the physical and chemical properties of the lagoon. The residence time of reef water in the lagoon was estimated to be 15–50 days based on published values for similar types of atolls in the western Pacific Ocean [Suzuki and Kawahata, 2003]. Because our sampling intervals (weekly or biweekly) were shorter than the residence time of reef water, our data should capture any changes in reef water chemistry associated with ambient water intrusion. There are no reports of coral bleaching or other major disturbances (e.g., fishing and pollution) in Chuuk lagoon [Houk *et al.*, 2015].

2.2. Seawater Sampling and Measurements of Seawater Carbonate Parameters

Sampling was performed in the morning (9–10 A.M.) in the northeastern part of the lagoon (7.27°N and 151.53°E), which has a depth of 23 m and is representative of the Chuuk lagoon (Figure 1c). From April 2008 to August 2013, approximately 500 ml of surface seawater (<1 m depth) was sampled weekly. For the subsequent period (September 2013 to August 2014), seawater samples were taken biweekly. Sampled seawater was immediately treated with mercuric chloride to prevent biological activity and then stored in sealed borosilicate bottles until analysis [Dickson *et al.*, 2007].

The A_{T} and C_{T} concentrations in all samples were determined using potentiometric and coulometric titration, respectively, both of which are integrated in the VINDTA system (Marianda, Kiel, Germany). For A_{T} measurements the samples were titrated with HCl (~ 0.2 M) to a pH of ~ 3.0 ; during the titration the volume of acid added was recorded at an average of 25 pH values. Values of A_{T} were determined from resulting titration data using a non-linear least squares approach [Millero *et al.*, 1993; Dickson *et al.*, 2007]. The accuracy of the measured A_{T} and C_{T} concentrations during the study period was checked daily by titrating reference seawater samples with known values of A_{T} and C_{T} prepared and distributed by A. Dickson, Scripps Institution of Oceanography. Any discrepancies between the measured and certified values for the references materials were applied to the measured values obtained for reef water samples analyzed on that day. After this adjustment, the precisions of the measured A_{T} and C_{T} concentrations determined during the study period were $\pm 1.7 \mu\text{mol kg}^{-1}$ and $\pm 2.1 \mu\text{mol kg}^{-1}$, respectively (Figure S1, supporting information). Phytoplankton biomass in the Chuuk lagoon remained low (<1 mg chl a m^{-3} derived from Moderate Resolution Imaging Spectroradiometer; <http://modis.gsfc.nasa.gov/>), suggesting that the contributions of phytoplankton cells and fresh dissolved organic matter produced by phytoplankton to A_{T} were not significant ($<1 \mu\text{mol kg}^{-1}$) [Kim *et al.*, 2006; Kim and Lee, 2009].

Other carbonate parameters (pH, pCO_2 , and Ω_{AR}) were calculated from A_{T} and C_{T} using a seawater thermodynamic model that includes the carbonic acid dissociation constants of Mehrbach *et al.* [1973], as refit by Dickson and Millero [1987] and other ancillary thermodynamic constants, including the dissociation constants for boric and phosphoric acids [Millero, 1995]. This set of thermodynamic constants has been shown to be valid when working with both laboratory and field data [Lee *et al.*, 1996, 2000; McElligott *et al.*, 1998; Lueker *et al.*, 2000; Millero *et al.*, 2002, 2006].

The A_{T} and C_{T} values of the ambient water surrounding the Chuuk lagoon were not measured directly, but rather estimated. A_{T} was estimated using an A_{T} parameterization expressed in terms of salinity and temperature [Lee *et al.*, 2006] and measured salinity and temperature for Chuuk reef water, which agreed well with measured values for ambient water collected within the 5° latitude \times 10° longitude area surrounding the Chuuk lagoon during our observational period (Figure S2). C_{T} values for the ambient water were estimated from pCO_2 values of seawater collected as part of the Surface Ocean CO_2 Atlas project, between 2008 and 2013, from the area (5°N – 10°N and 150°E – 160°E) that includes the Chuuk lagoon [Bakker *et al.*, 2014] (Figure S2c) and the estimated A_{T} using the thermodynamic model described above. Note that, because the seasonal pCO_2 data coverage for 2012 is greater than that for 2013 (see Figure S2), ambient water pCO_2 values for 2014 were estimated by adding 2 times ($3.0 \mu\text{atm year}^{-1}$) the annual pCO_2 increase to the 2012 annual cycle of pCO_2 . The estimated A_{T} and C_{T} values were consistent to $\pm 5 \mu\text{mol kg}^{-1}$ with the values measured along the 148°E section in 2010 as part of the CO_2 /Climate Variability Repeat Hydrography Program.

3. Results and Discussion

3.1. Seawater Carbonate Parameters in the Chuuk Reef and Ambient Environment

The Chuuk lagoon time series data show distinct seasonal oscillations in all carbonate parameters (Figures 2a–2d). In particular, both A_{T} and C_{T} reached their annual maxima in January through March, decreasing gradually

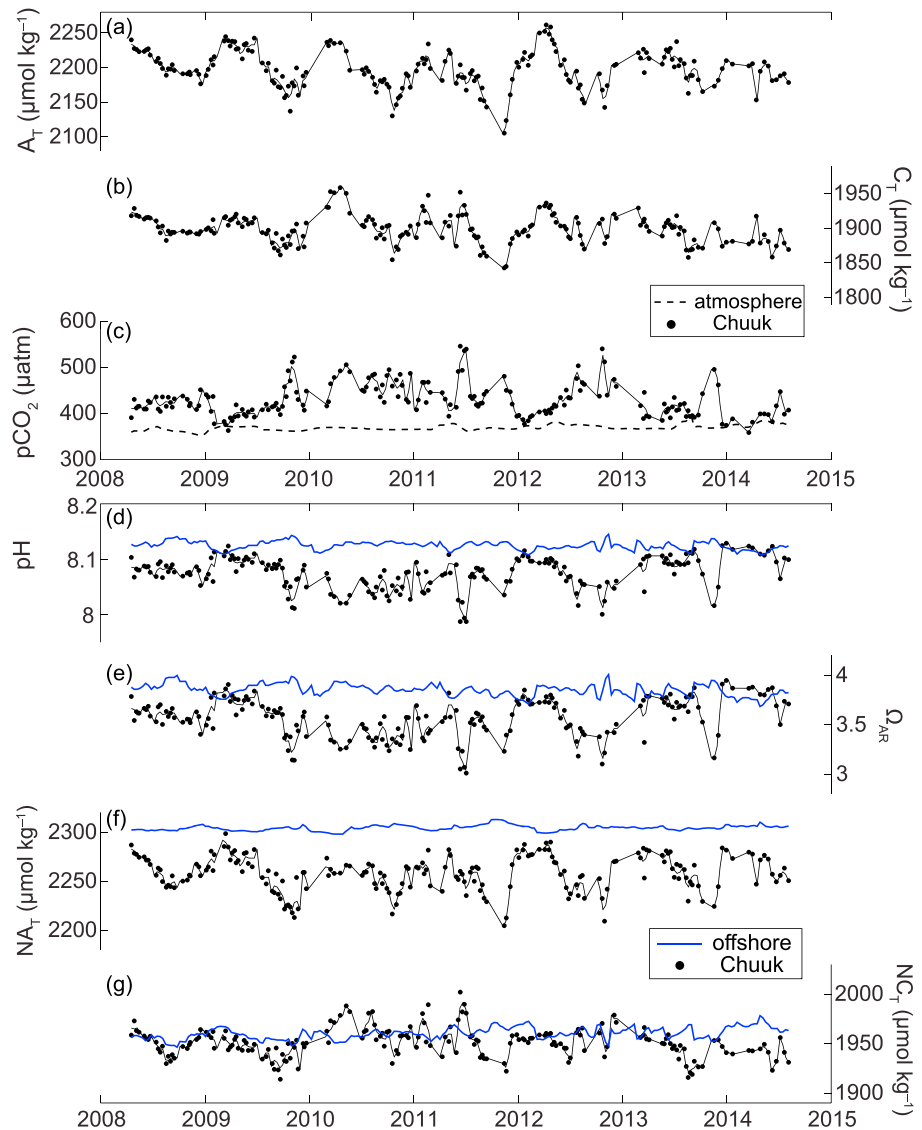


Figure 2. Time series of measured (a) A_T , (b) C_T , (c) pCO_2 , (d) pH, (e) Ω_{AR} , (f) NA_T , and (g) NC_T on the Chuuk lagoon. pCO_2 and Ω_{AR} values were calculated at in situ temperature and salinity and pH values were calculated at 25°C. We removed variations in A_T and C_T associated with salinity variations by normalizing the A_T and C_T values ($NA_T = A_T \times 35/S$ and $NC_T = C_T \times 35/S$). The solid circles indicate measured values and the solid lines indicate the best fits of monthly means. The dashed line in Figure 2c represents the atmospheric value of pCO_2 and the blue lines in Figures 2d–2g represent ambient water values for pH, Ω_{AR} , NA_T , and NC_T .

until reaching their minima in November–December. A_T oscillations were more dramatic and clearer than oscillations in C_T . Such seasonal patterns were also evident in the salinity (S)-normalized A_T and C_T (NA_T or $NC_T = A_T$ or $C_T \times 35/S$) (Figures 2f and 2g), but the patterns were less clear. More interestingly, pH, Ω_{AR} , NA_T , and NC_T all remained lower than the ambient water values (blue lines in Figures 2d–2g) throughout the observation period, whereas pCO_2 was almost always higher than atmospheric pCO_2 (Figure 2c). The large reductions (20–90 $\mu mol kg^{-1}$) from ambient values of NA_T suggest that net calcification has been a major driver of changes in the carbonate system of Chuuk reef water (Figure 2f). This conclusion was further strengthened by companion Ω_{AR} data (Figure 2e), showing that Ω_{AR} in reef water remained lower than the ambient values during the same period. Such strong calcification signals were less discernible for NC_T (Figure 2g), probably because variations in NC_T were influenced not only by net calcification but also by respiration.

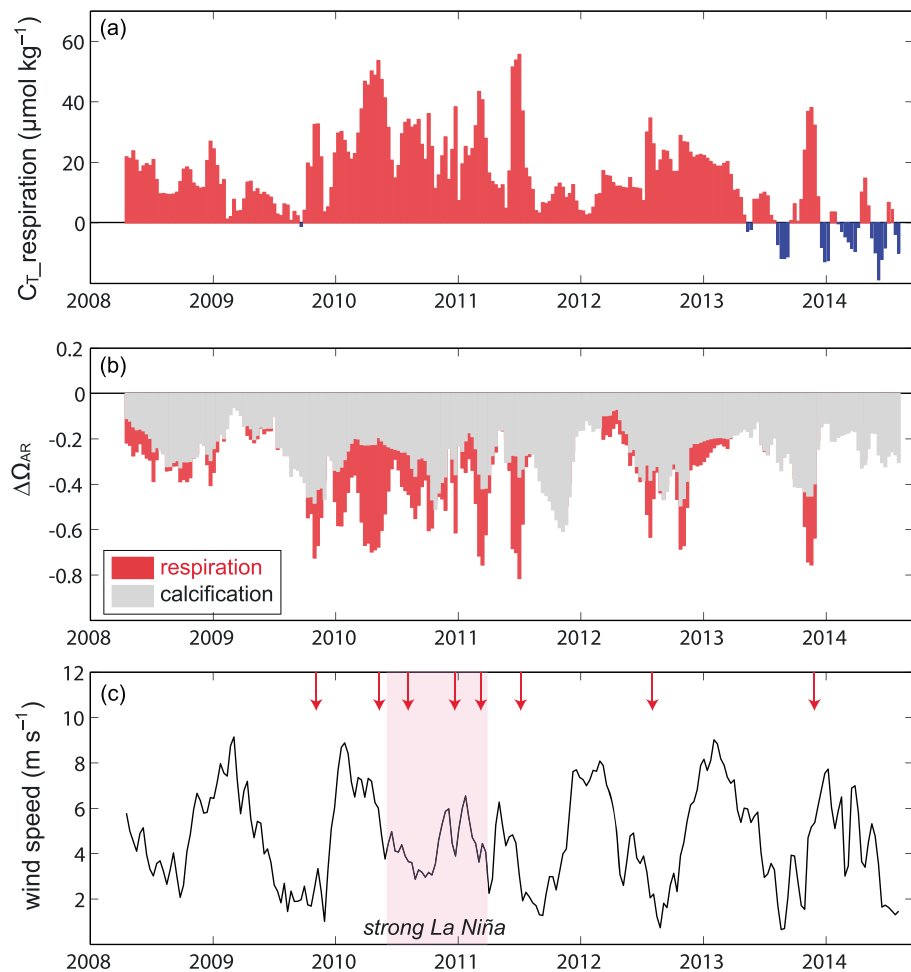


Figure 3. Time series of (a) respiration-driven C_T ($C_{T-respiration} = NC_{T-reef\ water} - NC_{T-offshore} - (NA_{T-reef\ water} - NA_{T-offshore})/2$). $C_{T-respiration}$ indicate the relative strengths of respiration and photosynthesis; positive values (red) indicate respiration exceeding photosynthesis, and negative values (blue) indicate photosynthesis exceeding respiration. Time series of (b) $\Delta\Omega_{AR}$ decreases resulting from calcification (gray) and respiration (red); and (c) wind speeds obtained from NCEP Reanalysis data. Vertical red arrows in Figure 3c indicate periods during which $C_{T-respiration}$ was greater than $30\ \mu\text{mol kg}^{-1}$.

Over the 6 year observational period, the observed rate of increase for ambient water $p\text{CO}_2$ ($1.50\ \mu\text{atm yr}^{-1}$) was similar to the rate of $1.88\ \mu\text{atm yr}^{-1}$ observed at ALOHA station (22°N and 148°W) for the period 1988–2007 [Dore et al., 2009]. Seasonal variations in the $p\text{CO}_2$ values of ambient water surrounding the Chuuk lagoon were only half the magnitude observed in a similar oligotrophic warm region (e.g., Bermuda Atlantic Time series site; 32°N and 64°W) [Takahashi et al., 2014]. The small variations in $p\text{CO}_2$ in waters surrounding the Chuuk lagoon can be attributed to the small variations in SST ($<2^\circ\text{C}$), which are the primary driver of $p\text{CO}_2$ changes in the oligotrophic ambient water surrounding the Chuuk lagoon.

3.2. Effects of Calcification, Photosynthesis, and Respiration on the Carbonate Chemistry of Reef Water

To estimate the contribution of photosynthesis and respiration ($C_{T-respiration}$) to the total change in C_T , we first estimated the change in C_T due to calcification ($[NA_{T-reef\ water} - NA_{T-offshore}]/2$) and then subtracted that from the total change in C_T relative to ambient conditions. Thus, a positive value indicates a contribution due to respiration (red shaded area in Figure 3a), and a negative value indicates an uptake of C_T due to photosynthesis (blue shaded area in Figure 3a). Given that $p\text{CO}_2$ of Chuck lagoon remained higher than atmospheric $p\text{CO}_2$, there is almost always a loss of C_T to the atmosphere through air-sea gas exchange (Figure 2c). Therefore, our estimates of $C_{T-respiration}$ should be considered somewhat lower than actual values. $C_{T-respiration}$ was found to be highest ($>30\ \mu\text{mol kg}^{-1}$) during much of 2010–2011 (Figure 3a).

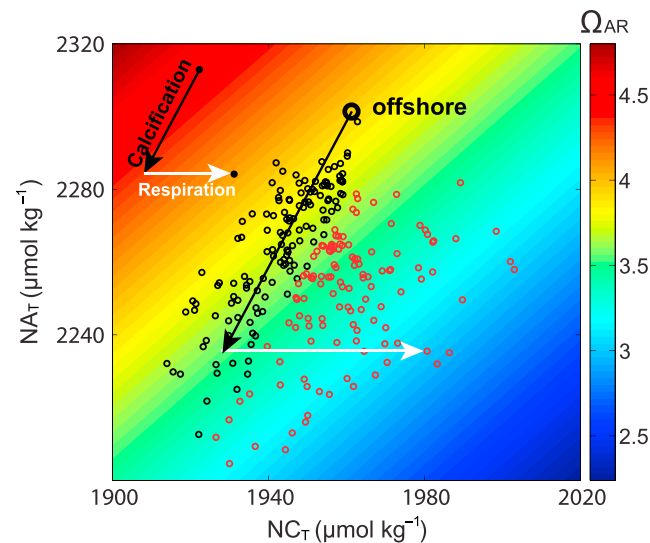


Figure 4. Plots of NA_T and NC_T overlaid with Ω_{AR} . Solid black arrows indicate changes in NA_T and NC_T due to calcification and solid white arrows indicate changes in NC_T due to respiration.

was close to that of seawater in equilibrium with atmospheric pCO_2 (550 μatm), the projected level for the year 2050 under the RCP8.5 emission scenario [Moss *et al.*, 2008].

3.3. Effect of Ambient Water Intrusion on the Carbonate Chemistry of Reef Water

Another and probably equally important process was the intrusion of ambient seawater surrounding the Chuuk reef system. These intrusions were estimated using NA_T anomalies between reef water and surrounding open water, with smaller NA_T anomalies indicating greater ambient water intrusion and mixing with reef water. The magnitude of ambient water intrusion was clearly evident when NA_T (less so NC_T) in the reef water approached ambient values, particularly between December and March. The NA_T increase during this time window could have also been caused by the enhancement of $CaCO_3$ dissolution, but this is unlikely given the high saturation states of the reef water during this transition season ($\Omega_{AR} > 3.0$).

The intrusion of ambient water from the surrounding open Pacific Ocean was generally greater during periods when the northeast trade winds are strongest, i.e., December and March. By contrast, intrusion of ambient water was low when wind speeds were reduced, coinciding with a decrease in reef water pH. Findings during 2010–2011 highlight the correlation between weakened winds and reef water carbonate chemistry. For much of 2010–2011, during which the Southern Oscillation Index (SOI) was among the highest (SOI > 20) ever recorded since the 1950s (SOI values from the Bureau of Meteorology in Australia are available at <http://www.bom.gov.au/climate/current/soihtm1.shtml>). The weakening of wind speed was widespread in much of the western tropical Pacific Ocean (blue in Figure 1b). In the Chuuk lagoon, seasonal wind speeds were approximately 30% lower than during other years (Figure 3c). Thus, the intrusion of ambient water into the Chuuk reef system (as indicated by the NA_T anomaly, Figure 2f) was weaker than during other years.

3.4. Drivers of Reef Water Ω_{AR} Change

The $NA_T - NC_T$ diagram emphasizes the relative strengths of net community calcification (NCC) and net community production (NCP) when determining the reef water carbonate system (A_T , C_T , and Ω_{AR}) during the study period. The slope of 2 of the plot of NA_T versus NC_T for the Chuuk lagoon indicates that NCC was a major driver of change in the carbonate system of reef water (black circles in Figure 4). In contrast, NCP was a minor contributor, being important only under conditions of weak intrusion of ambient water. During the periods of weak intrusion (e.g., much of 2010–2011 and from summer 2012 to spring 2013), the NA_T and NC_T data deviated considerably from the slope of 2, falling on the right side of this slope (red circles in Figure 4). A greater deviation of C_T from a slope of 2 indicates a greater respiration-driven C_T . Under

Another large signal in respiration-driven C_T was observed between the summer of 2012 and the spring of 2013. During these two periods (2010–2011 and 2012–2013), $C_{T-respiration}$ contributed 30–50% of the total decrease in Ω_{AR} (~0.6); the remainder was attributed to the decrease in A_T because of calcification (Figure 3b). In other years, the calcification-driven A_T decrease was a major driver of change in reef water Ω_{AR} . It is important to note that higher $C_{T-respiration}$ corresponds to higher surface pCO_2 (Figure 2c).

The ~0.6 reduction in Ω_{AR} observed in 2010 and 2011 may have decreased coral calcification rates by 9–40%, according to a recent synthesis of calcification studies [Chan and Connolly, 2013]. The reduced Ω_{AR} , 0.6 lower than in present-day ambient water,

conditions of weak intrusion of ambient water, the benthic respiration process was important in contributing to the decrease in Ω_{AR} in Chuuk reef water.

The $NA_T - NC_T$ slope observed during the 6 year period in the Chuuk lagoon is near the high end of the reported values, ranging from 0.26 to 2.02 [Suzuki and Kawahata, 2003; Andersson et al., 2014; Lantz et al., 2014]. The $NA_T - NC_T$ slope of a coral reef system has important implications for the response of that system to anthropogenic ocean acidification. Depending on major reef metabolic processes, including calcification, organic matter production, and respiration, reef water Ω_{AR} will follow distinct paths that differ from those predicted from the oceanic uptake of anthropogenic CO_2 alone. If the decrease in C_T due to the net production of organic matter were greater than the C_T decrease resulting from calcification ($\Delta A_T / \Delta C_T < 1$; Case 1 following the definition given in Andersson and Gledhill [2013]), then the former would partially offset the decrease in Ω_{AR} resulting from calcification and the oceanic uptake of anthropogenic CO_2 [Andersson and Gledhill, 2013]. In contrast, if the decrease in C_T resulting from calcification were greater than the C_T decrease resulting from net production of organic matter ($\Delta A_T / \Delta C_T > 1$; Case 2 following the definition given in Andersson and Gledhill [2013]), then all three processes (i.e., calcification, respiration, and oceanic uptake of anthropogenic CO_2) would act to lower the pH of reef water [Andersson and Gledhill, 2013]. Therefore, Case 2 reef water systems such as Chuuk lagoon will be particularly vulnerable during the coming decades. Although the carbonate chemistry of relatively few reefs has only rarely been measured over time periods adequate to classify them as either Case 1 or Case 2, several western Pacific coral reef systems, including the Palau barrier reef, Majuro atoll, and Northern and Southern Great Barrier Reefs, appear to be Case 2 reefs [Kawahata et al., 1997, 1999; Suzuki and Kawahata, 2003]. More studies of a wide range of coral reefs are required to identify the dominant reef water carbonate chemistry of global coral reef systems.

4. Conclusions and Implication

During the study period, the measured NA_T and Ω_{AR} in Chuuk reef water were considerably lower than the values in the surrounding ambient water. The large reductions ($20\text{--}90 \mu\text{mol kg}^{-1}$) in NA_T compared to ambient values point to net calcification as a major driver of changes in the carbonate system of the Chuuk lagoon. However, during La Niña conditions, NC_T values were increased by $20\text{--}50 \mu\text{mol kg}^{-1}$. Under such conditions, the wind speed is lower, thus reducing the intrusion of ambient water from the surrounding open water and increasing the residence time of reef water. Under the resulting longer residence time conditions, reef water respiration was the only process that increased NC_T . Calcification and greater production of CO_2 during La Niña phase of the ENSO reinforced each other, markedly lowering the pH of reef water. More alarmingly, the lowered Ω_{AR} condition persisted for more than 6 months, which may further lower the rate of calcification, thereby jeopardizing the overall health of the coral reef habitat. The model projections of weakening trade winds in the equatorial Pacific Ocean during coming centuries [Collins et al., 2010] suggest that weak intrusion of ambient water will occur more frequently, thereby intensifying the associated reduction in Ω_{AR} in the coral-rich western equatorial Pacific Ocean.

The present study discloses a strong link between reef water carbonate chemistry and large-scale climatic variations. These findings can be extended throughout the coral-rich western tropical Pacific Ocean, where ambient water intrusion into reef systems is influenced by wind speed variations closely associated with variations in ENSO events. The results also have broader implications, in that they suggest that temporal variations in the carbonate chemistry of semienclosed reef systems with limited water exchange, ranging from seasons to decades, may be controlled more by ambient water intrusion than by oceanic uptake of anthropogenic CO_2 .

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