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2 **Could artificial ocean alkalization protect tropical coral** 3 **ecosystems from ocean acidification?**

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7 **Abstract:** Artificial ocean alkalization (AOA) is investigated as a method to mitigate local ocean
8 acidification and protect tropical coral ecosystems during a 21st century high CO₂ emission scenario.
9 Employing an Earth system model of intermediate complexity, our implementation of AOA in the
10 Great Barrier Reef, Caribbean Sea and South China Sea regions, shows that alkalization has the
11 potential to counteract expected 21st century local acidification in regard to both oceanic surface
12 aragonite saturation Ω and surface pCO₂. Beyond preventing local acidification, regional AOA,
13 however, results in locally elevated aragonite oversaturation and pCO₂ decline. A notable consequence
14 of stopping regional AOA is a rapid shift back to the acidified conditions of the target regions. We
15 conclude that artificial ocean alkalization may be a method that could help to keep regional coral
16 ecosystems within saturation states and pCO₂ values close to present-day values even in a
17 high-emission scenario and thereby might “buy some time” against the ocean acidification threat, even
18 though regional AOA does not significantly mitigate the warming threat.

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25 1. Introduction

26 Anthropogenic CO₂ invades the ocean and thereby perturbs ocean chemistry, this phenomenon is
27 also known as “ocean acidification” (e.g. Caldeira and Wickett 2003, Feely *et al* 2004). If CO₂
28 emissions continue to increase and the ocean continues to become more acidic these changes will
29 further affect the ambient saturation state of aragonite (described by aragonite Ω). Since calcification,
30 which is a crucial skeleton building process for most stony corals, is considered to be highly sensitive
31 to ambient aragonite Ω , coral calcification is likely to become inhibited in the future (Gattuso *et al*
32 1998, Langdon and Atkinson 2005). Stony coral reefs sustain the most diverse ecosystems in the
33 tropical oceans, and the coral-supported tropical fish (Munday *et al* 2014), coralline algae (McCoy and
34 Ragazzola 2014), echinoderms (Dupont *et al* 2010), molluscs (Gazeau *et al* 2007), crustaceans
35 (Whiteley 2011), and corals themselves (Kleypas *et al* 1999a, Hoegh-Guldberg *et al* 2007, Cao and
36 Caldeira 2008, Crook *et al* 2011, Meissner *et al* 2012a) are expected to face difficulties in adapting to
37 future ocean conditions in coming decades because of both ocean acidification itself and the loss of the
38 reef structure. A potential loss of coral reefs and their ecosystems may also have a direct impact on
39 coastal resources and services (Brander *et al* 2009). Besides the threat from ocean acidification coral
40 reefs face a number of other significant threats such as coral bleaching, which is triggered by
41 persistent heat stress and is thought to be one of the most serious climate change related threats
42 (Hoegh-Guldberg 1999, Cooper *et al* 2008, De’ath *et al* 2009, Frieler *et al* 2012, Caldeira 2013)..

43 Since efforts to mitigate global warming and ocean acidification by reducing emissions have, up
44 to now, been unsuccessful in terms of a significant reduction in the growth of atmospheric CO₂
45 concentrations, there has been growing interest in climate engineering (CE) to mitigate or prevent
46 various consequences of anthropogenic climate change (Crutzen 2006, Schuiling and Krijgsman 2006,
47 Oeschlies *et al* 2010). For example, several modelling studies have examined “Artificial Ocean
48 Alkalinization (AOA)” which modifies ocean alkalinity. These studies simulated the use of alkalizing
49 agents such as olivine (a Mg-Fe-SiO₄ mineral) (Köhler *et al* 2010, 2013, Hartmann *et al* 2013),
50 calcium carbonate (Caldeira and Rau 2000, Harvey 2008), or calcium hydroxide (Ilyina *et al* 2013a,
51 Keller *et al* 2014) to elevate the ocean’s alkalinity to increase CO₂ uptake and mitigating ocean
52 acidification. While these simulations suggested that AOA could potentially be used to mitigate global
53 warming and ocean acidification to some degree, some studies also suggested that deploying AOA at a
54 global scale may face prohibitive logistical and economical constraints and could possibly cause
55 undesired side effects (Renforth *et al* 2013, Keller *et al* 2014).

56 In this paper we use Earth system model simulations of regional AOA to investigate the potential
57 of AOA to protect specific stony coral reef regions against ocean acidification. We also investigate
58 possible environmental side effects of AOA and possible regional differences in effectiveness or
59 undesired side effects. The model simulations show AOA could mitigate ocean acidification in our

60 investigated coral reef regions, albeit at substantial economic costs and with the termination risk of a
61 rapid return to acidified conditions after the stop of local AOA.

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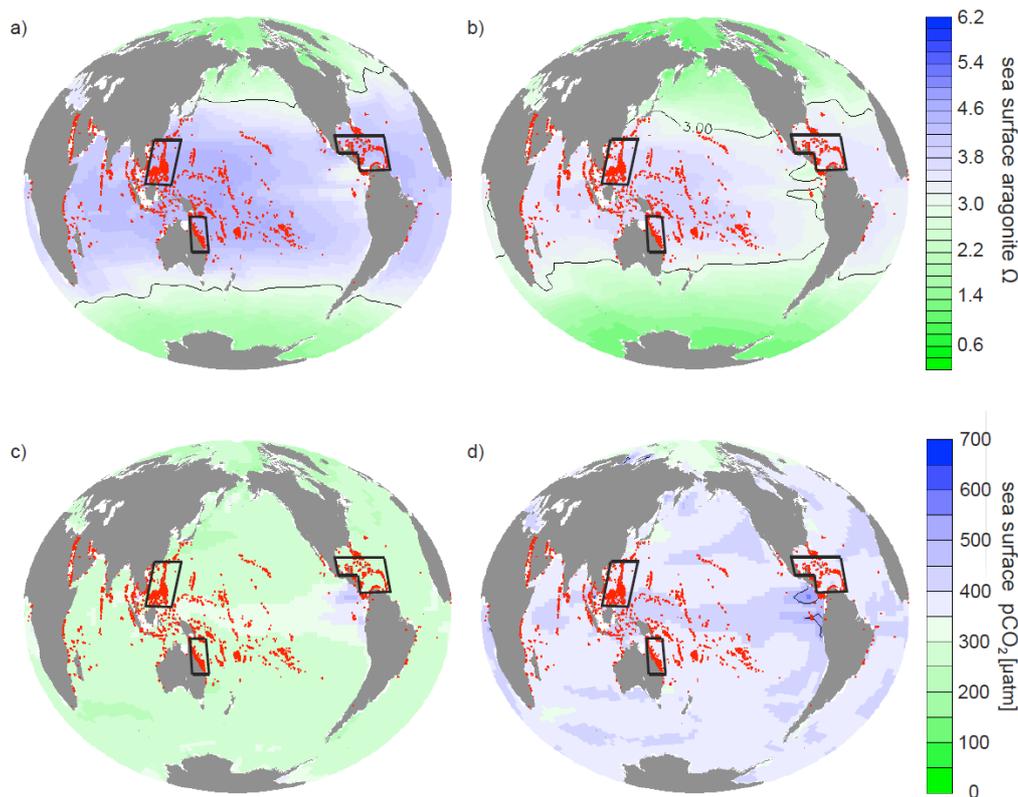
63 **2. Methods**

64 We simulated calcium hydroxide ($\text{Ca}(\text{OH})_2$) based AOA in the Great Barrier Reef (GB,
65 $9.0^\circ\text{S}\sim 27.0^\circ\text{S}$, $140.4^\circ\text{E}\sim 154.8^\circ\text{E}$, an area of $1.7 \times 10^6 \text{ km}^2$), the Caribbean Sea (CS, $10.8^\circ\text{N}\sim 27^\circ\text{N}$,
66 $68.4^\circ\text{W}\sim 93.6^\circ\text{W}$, an area of $3.9 \times 10^6 \text{ km}^2$) and the South China Sea (SC, $0^\circ\text{N}\sim 23.4^\circ\text{N}$,
67 $104.4^\circ\text{E}\sim 129.6^\circ\text{E}$, an area of $5.2 \times 10^6 \text{ km}^2$) (figure 1) using the University of Victoria Earth System
68 Climate Model (UVic) version 2.9. These areas contain some of the world's most abundant coral reefs
69 (<http://www.reefbase.org/>) and are large enough to be addressed by the UVic model. From the data
70 obtained from ReefBase (<http://www.reefbase.org/>), we found that from a total of 10,048 coral reef
71 locations, 3323 are located in the Great Barrier Reef box, 601 in the Caribbean Sea box, and 2060 in
72 the South China Sea box. Altogether 5984 reef points are included in our three regions, which is more
73 than half of the global coral reef locations collected from ReefBase.

74 The UVic model consists of an energy-moisture balance atmospheric component, a 3D
75 primitive-equation oceanic component that includes a sea-ice sub-component, and a terrestrial
76 component (Weaver *et al* 2001, Meissner *et al* 2003). Wind velocities are prescribed from
77 NCAR/NCEP monthly climatological data. Accordingly, UVic does not feature decadal
78 ocean-atmosphere oscillations, like ENSO. The model has a spatial resolution of $3.6^\circ \times 1.8^\circ$ with 19
79 vertical layers in the ocean. The global carbon cycle is simulated with air-sea gas exchange of CO_2 and
80 marine inorganic carbonate chemistry following the Ocean Carbon-Cycle Model Intercomparison
81 Project Protocols (Orr *et al* 1999). The inorganic carbon cycle is coupled to a marine ecosystem
82 model that includes phytoplankton, zooplankton, detritus, the nutrients nitrate and phosphate, and
83 oxygen (Keller *et al* 2012). The model has been evaluated in several model intercomparison projects
84 (Eby *et al* 2013, Zickfeld *et al* 2013, Weaver *et al* 2012), and shows a reasonable response to
85 anthropogenic CO_2 forcing that is well within the range of other models. In order to illustrate that our
86 model is robust in reproducing general ocean circulation and chemistry, we validate our model against
87 GLODAP (Global Ocean Data Analysis Data Project) v1.1 data for ocean total alkalinity and oceanic
88 dissolved inorganic carbon (Key *et al* 2004) (figures S1 and S2 in supplementary materials), SOCAT
89 (Surface Ocean CO_2 Atlas) data (Bakker *et al* 2014, Landschützer *et al* 2014) for sea surface pCO_2
90 (figure S3), and WOA (World Ocean Atlas) 2013 data for sea surface temperature (figure S4). The
91 validation illustrates that UVic can generally reproduce the global patterns of surface ocean alkalinity
92 and dissolved inorganic carbon as well as sea surface pCO_2 . UVic's performance in reconstructing sea
93 surface temperature (SST) is also generally good, especially in regions where AOA is implemented in
94 our study with less than a 0.8°C model-data misfit. Overall, the model-data differences displayed by

95 the UVic model are well within the range data-error bonds from CMIP5 model simulations (Wang *et*
96 *al* 2014, Ilyina *et al* 2013b, Jungclaus *et al* 2013).

97 The model was spun-up for 10,000 years under pre-industrial atmospheric and astronomical
98 boundary conditions. From year 1800 to 2005 the model was forced with historical fossil fuel and
99 land-use carbon emissions. Then, from the year 2006 onwards the Representative Carbon Pathway 8.5
100 (RCP 8.5) anthropogenic CO₂ emission scenario forcing was used (Meinshausen *et al* 2011). CO₂ is
101 the only greenhouse gas taken into account. Continental ice sheets, volcanic forcing, and astronomical
102 boundary conditions were held constant to facilitate the experimental set-up and analysis.



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105 **Figure 1.** Annual mean surface aragonite Ω and pCO₂ simulated by the UVic model control run
106 without regional artificial ocean alkalization (AOA) for preindustrial (a, c) and 2020 (b, d). AOA
107 experimental regions are marked by black boxes. Coral Reef locations are marked in cyan.

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109 Ca(OH)₂ based AOA is simulated in an idealized manner by increasing surface alkalinity (Keller
110 *et al* 2014). The rationale behind this method is that dissolving one mole of Ca(OH)₂ in seawater
111 increases total alkalinity by 2 moles (Ilyina *et al* 2013a). We simulate Ca(OH)₂-based AOA by
112 homogeneously and continuously adding alkalinity to the upper 50m of the targeted regions. In the
113 following, we therefore use the term “lime addition” to refer to our simulated Ca(OH)₂ addition.

114 Directly simulating individual reefs or corals is beyond our current model's capacity and we therefore
115 focus on AOA-induced impacts on regional and global marine chemistry. Also, we ignore the impact
116 of increasing water temperature on corals, which will accompany elevated levels of atmospheric CO₂
117 and would likely also have a detrimental impact on coral reefs.

118 We use a fixed threshold aragonite Ω to describe suitable stony coral habitats since most of
119 today's coral reefs are found in waters with ambient seawater aragonite Ω above a critical value
120 (Kleypas *et al* 1999b, Meissner *et al* 2012a, 2012b, Ricke *et al* 2013). However, this approach involves
121 some uncertainties (Kleypas *et al* 1999b, Guinotte *et al* 2003) due to the neglect of seasonal and
122 diurnal Ω fluctuations, species variety, and species ability to adapt. Critical coral habitat threshold
123 values of ambient aragonite Ω ranging from $\Omega = 3$ (Meissner *et al* 2012b), $\Omega = 3.3$ (Meissner *et al*
124 2012a), to $\Omega = 3.5$ (Ricke *et al* 2013) have been used in recent climate change studies, acknowledging
125 that these represent regional mean values and that local reef-scale carbonate chemistry may display
126 large diurnal fluctuations also in healthy reefs. Ignoring sea surface temperature as a regulator of coral
127 reef habitats may be a further simplification (Couce *et al* 2013). We follow these earlier studies
128 and, in this paper, use an aragonite Ω threshold of 3 to determine whether or not seawater chemistry
129 with a region is suitable for stony corals.

130 A healthy coral ecosystem usually includes a multitude of both calcifying and non-calcifying
131 organisms. Aragonite Ω is commonly used to evaluate the impact of ocean acidification on marine
132 calcifying organisms. Nevertheless, ocean acidification can also affect non-calcifying organisms, e.g.
133 by reducing their metabolic rates (Rosa and Seibel 2008) or damaging their larval and juvenile stages
134 (Frommel *et al* 2011). Concerning non-calcifying organisms, often pCO₂ is employed as a metric to
135 evaluate impacts of ocean acidification. We therefore also consider how seawater pCO₂ will develop
136 under increasing atmospheric pCO₂ and continuous AOA. Without AOA, annual mean surface
137 seawater pCO₂ will follow atmospheric pCO₂ with some small time lag (e.g. Bates 2007). A
138 meta-study of resistance of different marine taxa to elevated pCO₂ (Wittmann and Pörtner 2013) found
139 that 50% of the species of corals, echinoderms, molluscs, fishes and crustaceans are negatively
140 affected if seawater pCO₂ reaches high levels (between 632 to 1,003 μatm) with many species, except
141 for crustaceans, also being significantly affected by pCO₂ levels between 500-650 μatm . Among the
142 studied species, 57 % of echinoderms and 50 % of molluscs were negatively affected by the lowest
143 levels of experimental pCO₂ manipulations. Since the loss of even one species, such as a keystone
144 species, could potentially be detrimental for reef health, we chose a relatively low threshold of 500
145 μatm pCO₂ (as an annual average) to determine whether or not conditions were suitable for
146 maintaining a healthy reef habitat. Moreover, by choosing a lower threshold we can better account for
147 any variability in pCO₂ that may not be well simulated by our model. However, we must acknowledge
148 that there are considerable uncertainties concerning such a threshold. Furthermore, these thresholds

149 can be modulated by other environmental factors (Manzello 2015) and may not be absolutely
 150 applicable in every reef location. To avoid unnecessary complexity, the thresholds for both $p\text{CO}_2$ and
 151 Ω are considered here in terms of regional and annual averages.

152 Four sets of model simulations were carried out (table 1), beginning at the start of the year 2020
 153 and ending at the end of the year 2099 of the RCP 8.5 emission scenario. Ensemble A is the control
 154 run (no AOA). In Ensemble B constant amounts of lime (from 1 to 10 Gt yr^{-1} with 1 Gt yr^{-1} increments)
 155 were added homogenously to each region. In Ensemble C we sought a solution where a linear increase
 156 of AOA over time ensured that our thresholds were met with a minimum lime addition, with the
 157 chosen rate of increase guided by the results from Ensemble B. Runs of Ensemble D are identical to
 158 those of Ensemble C, except for the fact that we stop AOA at the beginning of the year 2070 and
 159 continue the run without AOA until the end of the year 2099. This is to study the impact of a planned
 160 or unplanned stop of AOA.

161

Table 1. Description of simulated Artificial Ocean Alkalinization (AOA) experiments during RCP 8.5 CO_2 emissions scenario forcing

Experimental Ensemble	AOA starts in year	AOA ends in year	Lime addition (Gt yr^{-1})	Number of runs [†]
A (control)	-	-	0	1
B (constant addition)	2020	2099	1, 2, 3, 4, 5, 6, 7, 8, 9, and 10	10
C (optimal)	2050* or 2048**	2099	Linear increase with time	60
D (optimal/termination)	2050* or 2048**	2070	Linear increase with time	60

162 [†] in each region respectively

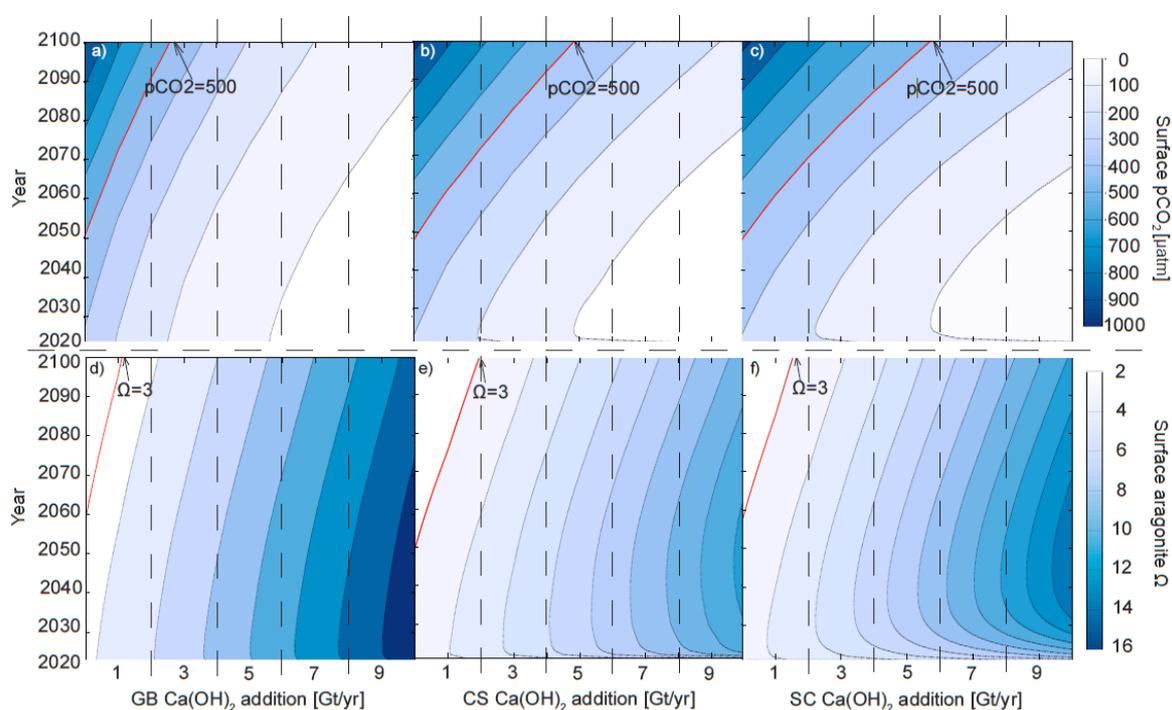
163 * Great Barrier Reef; ** Caribbean Sea and South China Sea

164

165 3. Results

166 In the control run, regionally averaged surface aragonite Ω drops below 3 in the Great Barrier
 167 Reef (GB) after year 2057, in the Caribbean Sea (CS) after year 2049, and in the South China Sea (SC)
 168 after year 2057 (figure 2). The mean $p\text{CO}_2$ threshold of 500 μatm is crossed in the GB at year 2050, in
 169 the CS at year 2048, and in the SC at year 2048. With constant AOA (Ensemble B) the thresholds are
 170 crossed at a later date or not at all, depending on the intensity of AOA. After an initial increase of Ω
 171 and decrease of $p\text{CO}_2$, respectively, the surface aragonite Ω declines and $p\text{CO}_2$ increases almost

172 linearly with time as ocean acidification intensifies because of the increasing invasion of atmospheric
 173 CO₂. The minimum amount of lime that is needed to prevent regionally averaged surface aragonite Ω
 174 from dropping below 3 before the end of year 2099 in these constant AOA simulations is 1.1 (GB), 1.9
 175 (CS), and 1.5 Gt yr⁻¹ (SC), respectively. In order to prevent regional annual-mean surface pCO₂ from
 176 exceeding 500 μ atm, the minimum amount of lime that is needed is always significantly larger, i.e. 2.5
 177 (GB), 4.9 (CS), and 5.7 Gt yr⁻¹ (SC), respectively. These results indicate that meeting the pCO₂
 178 threshold in our setup always requires a higher alkalinity addition than it does to meet the aragonite
 179 saturation threshold, thus in our particular case of combined pCO₂ and Ω thresholds, only the pCO₂



180 threshold needs to be considered.

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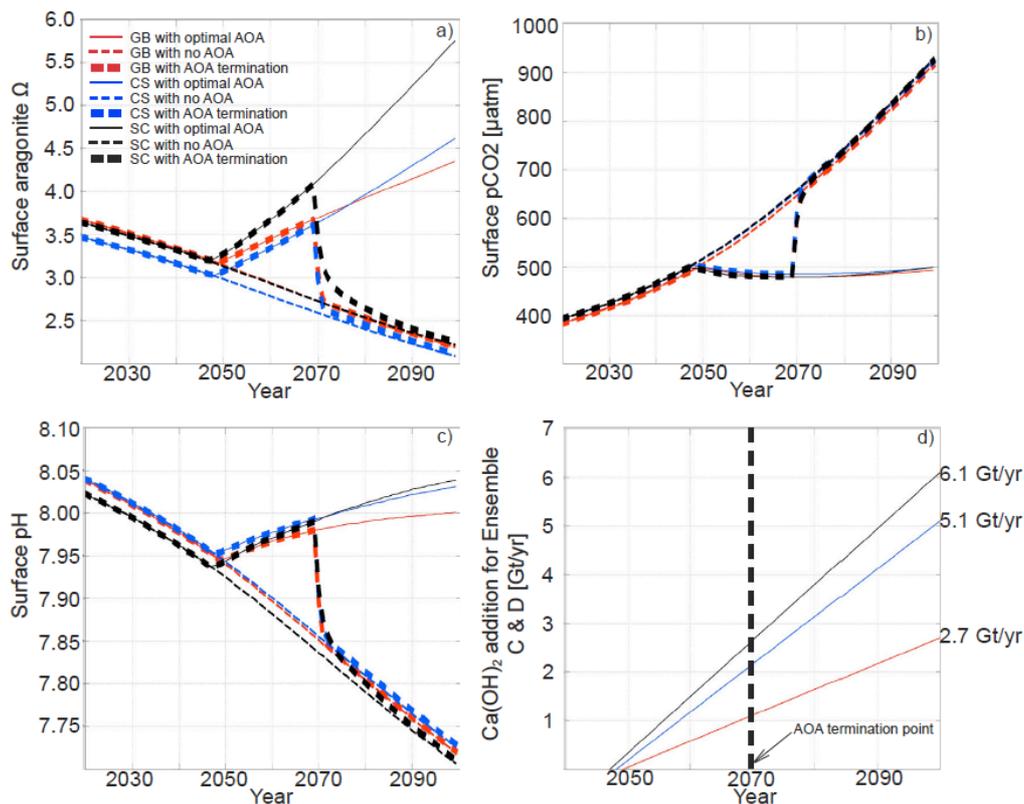
182 **Figure 2.** Regionally-averaged surface aragonite Ω and surface pCO₂ that occur in the Great Barrier
 183 Reef (a, d), Caribbean Sea (b, e), and the South China Sea (c, f) regions for the Ensemble A and B
 184 simulations as a function of time. Thresholds are highlighted by red isoclines.

185

186 Ensemble C includes a total of 60 model runs for each region that were initiated with output from
 187 the control run years 2050 (GB) and 2048 (CS and SC), respectively, which are the time points just
 188 before our chosen threshold values for surface pCO₂ was crossed in the respective experimental
 189 regions. Thereafter, simulated lime additions increase linearly from 0 Gt yr⁻¹ to a maximum addition in
 190 year 2099, which ranges from 2 to 7 Gt yr⁻¹ depending on the region (not shown; see figure 3(d) for

191 the "optimal" example). Of the 3 x 60 runs composing Ensemble C, our specific interest was in the
 192 runs ending at 2.7 (GB), 5.1 (CS) and 6.1 (SC) Gt lime per year (year 2100) since these "optimal" runs
 193 require the least time integrated amount of AOA to prevent our chosen thresholds from being crossed
 194 (figure 3(b)). In year 2099 of these runs we find surface aragonite Ω =4.3, 4.6, and 5.7 and surface
 195 pH=7.99, 8.03, and 8.04 in the GB, CS, and SC, respectively (figure 3(a)). That is, in order to prevent
 196 local seawater pCO₂ from increasing above our chosen threshold, one would have to accept a
 197 considerable increase in seawater Ω compared to the situation in 2020. In the year 2099, the
 198 region-averaged alkalinity additions are 42.6 mol m⁻² yr⁻¹ (GB), 34.9 mol m⁻² yr⁻¹ (CS) and 31.2 m⁻²
 199 yr⁻¹ (SC). This regional AOA leads to an additional global oceanic carbon uptake of ~15.36, 32.54, and
 200 35.41 Gt C for the GB, CS, and SC runs by the end of the year 2099, respectively.

201 Terminating regional AOA (Ensemble D) has a strong and rapid impact on surface aragonite Ω ,
 202 seawater pCO₂ and pH in the respective regions (figure 3). After termination the AOA related regional
 203 changes disappear on an annual timescale and quickly converge back to conditions very close to those
 204 of the control run.

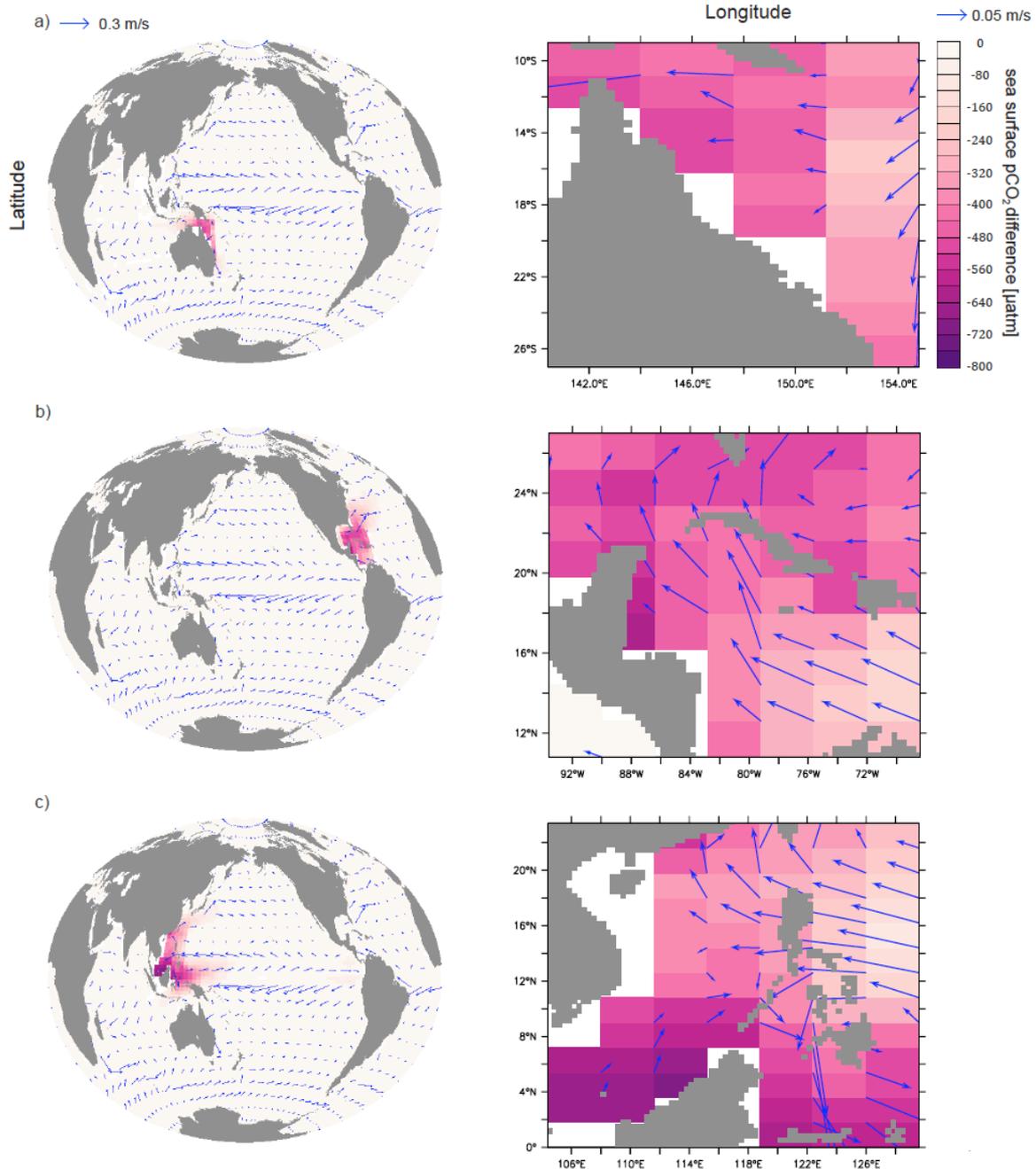


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 206 **Figure 3.** Comparison between the Great Barrier Reef, Caribbean Sea, and South China Sea regionally
 207 averaged annual surface aragonite Ω (a), seawater pCO₂ (b), and sea surface pH (c) values during the
 208 control (Ensemble A) and the "optimal" AOA simulations (single optimized simulation from

209 Ensembles C and D). Note that AOA ends in the year 2070 in the Ensemble D simulations. The
210 amount of lime needed for the “optimal” AOA implementation in year 2100 is labelled in (d).

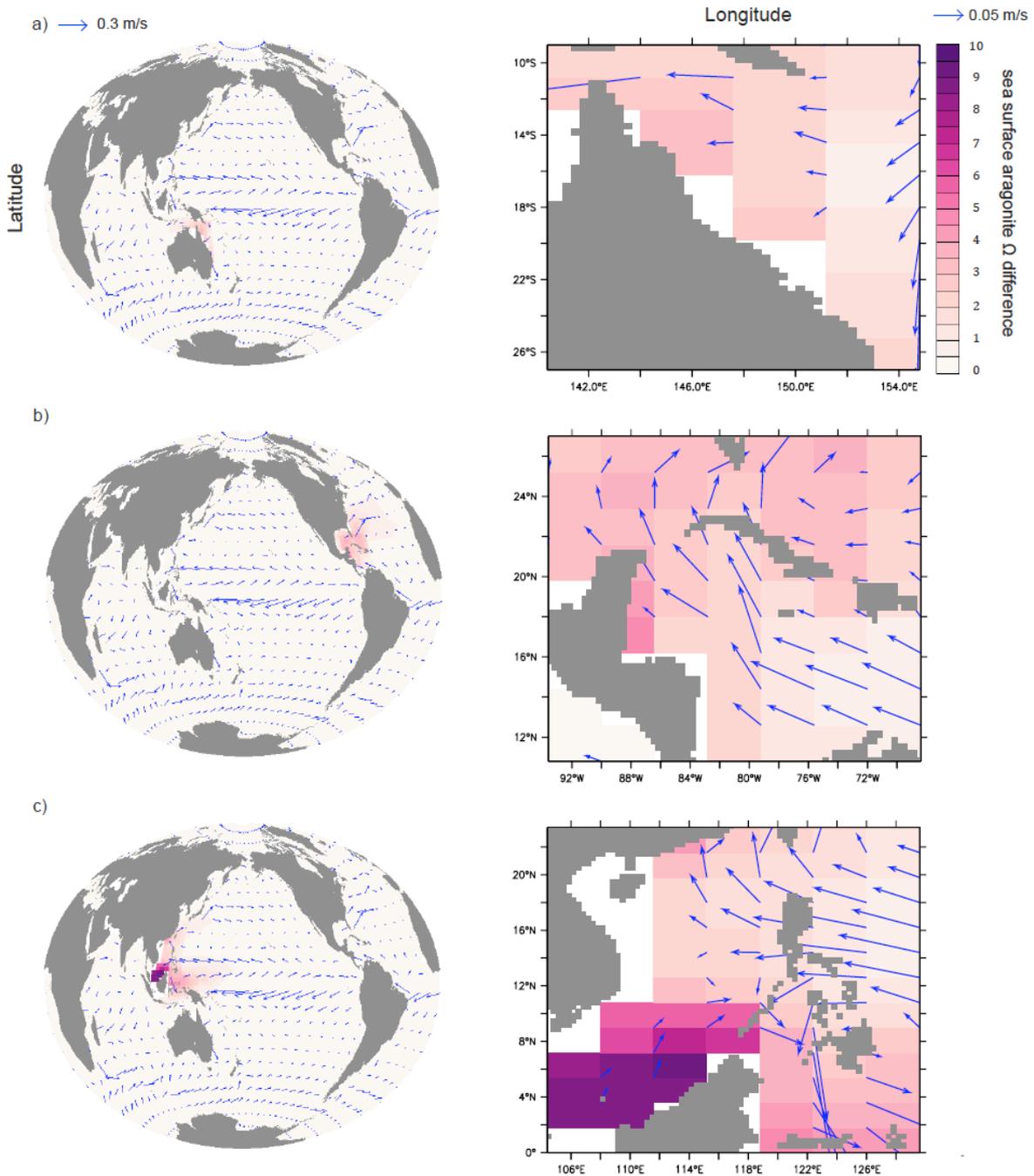
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212 Regional AOA also has effects on global ocean biogeochemistry (figures 4 and 5). Within a few
213 decades, AOA in the CS affects surface Ω and seawater $p\text{CO}_2$ in much of the western North Atlantic.
214 On the timescales considered, AOA in the GB region appears to be the most locally confined in our
215 runs, but nevertheless affects the coastal waters of Papua New Guinea and Indonesia. Overall,
216 however, remote effects are moderate compared with local impacts. Compared with the control run,
217 the optimal runs (Ensemble C) have annual regional surface $p\text{CO}_2$ partial pressures that are ~ 300 to
218 $800 \mu\text{atm}$ lower and an aragonite Ω that is of ~ 2.5 to 10 times higher in AOA regions compared to the
219 control run without AOA. At the same time, both the globally averaged increase in surface Ω and the
220 decrease in $p\text{CO}_2$ are moderately small (figure S5 in supplemental materials). Thus, in our optimal
221 AOA simulations, atmospheric CO_2 is drawn down by the end of 2099, relative to the control run, by
222 about 7 ppm for GB run, 15 ppm for CS run and 16 ppm for SC.



223

224 **Figure 4.** Simulated year 2099 surface pCO₂ differences between the optimal runs (Ensemble C) and
 225 the control run for the Great Barrier Reef (a), Caribbean Sea (b), and South China Sea (c) Each is
 226 shown with respect to the global impact (left) and the impact over the respective region where AOA is
 227 applied (right). Annual mean surface current velocities are marked as blue arrows.



228

229 **Figure 5.** Simulated year 2099 surface aragonite Ω differences between the optimal runs (Ensemble C)
 230 and the control run for the Great Barrier Reef (a), Caribbean Sea (b), and South China Sea (c). Each is
 231 shown with respect to the global impact (left) and the impact over the respective region where AOA is
 232 applied (right). Annual mean surface current velocities are marked as blue arrows.

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236 4. Discussion

237 From a marine biogeochemical perspective, our results indicate that regional AOA could
238 potentially be an effective means to mitigate regional ocean acidification. In our AOA simulations
239 (Ensemble B, C and D) the increase of surface seawater pCO₂ levels, as well as the reduction of local
240 pH and aragonite saturation states are all mitigated or even reversed in the targeted regions (figure 2, 3,
241 4). However, increasing surface ocean alkalinity also induces an additional uptake of CO₂. For the
242 optimal runs (Ensemble C), AOA modifies the oceanic DIC system (figure S6) leading to an increase
243 in both carbonate and bicarbonate ions. The increase of Ω and the carbonate ion concentration beyond
244 current or preindustrial levels in figure 3(a) may have unforeseen consequences in the real ocean as
245 elevated supersaturation may have biological impacts (Cripps *et al* 2013) or even cause the
246 spontaneous abiotic precipitation of CaCO₃. Like the biotically induced precipitation of CaCO₃, this
247 process would directly lead to an increase of pCO₂, i.e. constituting a negative feedback to intentional
248 alkalization. If spontaneous CaCO₃ precipitation due to elevated total alkalinity happens, this would
249 be detrimental as coral reefs are known to be sensitive to high levels of turbidity (Broecker and
250 Takahashi 1966, Roy and Smith 1971). Previous research by Renforth *et al* (2013) suggest to use an
251 optimum lime particle size of 80~100 since such particles can be fully dissolved in a typical surface
252 ocean with a depth less than 100 meters. For most tropical stony coral ecosystems, which are generally
253 within 100 meters of the surface, the direct addition of such particulate lime could affect water column
254 transparency and may even result in particles settling directly onto organisms. To minimize this side
255 effect, lime could be dissolved in seawater before adding it.

256 In addition to CO₂-system changes, AOA, if done with lime, will add calcium to the system. In
257 our optimum simulations (Ensemble C), the surface calcium concentration could be elevated by up to
258 0.16, 0.26, and 0.34 mmol/kg for the GB, CS, and SC respectively (figure S7 in supplementary
259 materials). Natural calcium behaves conservatively in the ocean and for a salinity of 35 the calcium
260 concentration is about 10.27 mmol/kg (Pilson 2013). The amount of calcium added during
261 alkalization is hence less than 4% of the background calcium concentration.

262 How well do our simulations reflect the real environmental conditions that coral reef ecosystems
263 might experience during a high CO₂ climate scenario (control run) and AOA deployment? An estimate
264 of potential impacts of model errors in simulated carbonate chemistry (table S1) suggests uncertainties
265 in the calculated regionally averaged alkalinity requirements of less than 10%. This result indicates
266 that the seawater chemistry simulated by UVic is acceptable for such an initial study of potential AOA.
267 Three limitations of our study, however, remain: First, the current model's coarse resolution does not
268 resolve small-scale physical processes like boundary currents, local upwelling and temperature

269 variability around reef archipelagos or in shallow lagoons (Meissner *et al* 2012a). Studies with higher
270 resolution models would be necessary to assess these local aspects. Second, most reported massive
271 coral bleaching incidents are associated with El-Niño years (Aronson *et al* 2002), which are not
272 resolved in our UVic model simulations driven by climatological winds. The observed
273 ENSO-associated variability in our studied regions is relatively weak, as revealed by an analysis of
274 historical SST and air-sea delta-pCO₂ records (supplementary figure S14 and S15). However, it is not
275 well known how ENSO variability will develop in the future (Collins *et al* 2010)(Guilyardi *et al* 2009)
276 and thus its impact on our study region will remain another uncertainty. Third, local calcification,
277 dissolution, photosynthesis and respiration within coral reefs affect local ocean chemistry and are not
278 in detail included in our model. For example, over a diurnal cycle the aragonite saturation state may
279 vary considerably, as has been observed at a coral reef off Okinawa, Japan where Ω ranged from 1.08
280 to 7.77 (Ohde and Hossain 2004). Similarly, seawater pCO₂ has been observed to vary between 420
281 μatm to 596 μatm during a 24 hour period (Dufault *et al* 2012). These large variations are due to
282 day-night fluctuations in carbon uptake and metabolism within coral reefs (Comeau *et al* 2012), which
283 our model cannot simulate. Because the ocean's uptake of anthropogenic CO₂ is associated with a
284 decrease in the ocean's buffering capacity, natural fluctuations of the carbonate system on seasonal
285 and diurnal scales are expected to increase (Riebesell *et al* 2009, Melzner *et al* 2012). Lacking the
286 small-scale variability in general, our simulations may underestimate the stress that might go along
287 with even stronger fluctuations in the future. However, since AOA would increase the regional ocean
288 buffering capacity, it could dampen future carbonate system fluctuations otherwise expected in a high
289 CO₂ emission world.

290 Our results also reveal different regional sensitivities of the AOA deployments. To stay within our
291 chosen mitigation guardrails (seawater pCO₂ < 500 μatm , Ω > 3 in the regional and annual means)
292 during the 21st century the GB requires the smallest amount of total lime input while the SC requires
293 the largest. These differences are largely due to the size difference of our studied regions. In the year
294 2099, the regional mean alkalinity additions are 42.6 mol m⁻² yr⁻¹ (GB), 34.9 mol m⁻² yr⁻¹ (CS) and
295 31.2 m⁻² yr⁻¹ (SC). These differences can be explained by a combination of local hydrography and
296 biogeochemistry. For example, the local surface alkalinity decline during the first year after AOA
297 termination in Ensemble D, i.e. between the beginning and end of year 2070 is largest in CS (80 mmol
298 m⁻³), intermediate for GB (70 mmol m⁻³) and smallest for SC (61 mmol m⁻³). Figure 3(b) reveals that
299 even though surface pCO₂ is almost identical in the three areas, the evolution of surface aragonite
300 saturation levels during AOA differs among the regions (figure 3(a)). For the same pCO₂ levels,
301 aragonite Ω in the SC increases more rapidly than in the CS and GB. This variation in carbonate
302 chemistry also leads to regionally different sensitivities to ocean acidification, which determines the
303 initiation and duration of AOA in Ensemble C.

304 The effectiveness of AOA can be described by the ratio between oceanic inventory changes of
305 DIC referenced to the control run and added total lime by the end of year 2099. The optimal runs of
306 Ensemble C show an effectiveness of 1.4 for GB, 1.5 for CS and 1.4 for SC, close to the value of 1.4
307 that we calculated based on data from Keller *et al* (2014). Compared to previous estimates of AOA
308 effectiveness that are above 1.6 (Renforth *et al* 2013), our slightly lower values can be explained by
309 the downward transport of added alkalinity on time scales shorter than the air-sea equilibration time of
310 CO₂ (figure S8 in supplementary material). This loss of alkalinity from the surface layer leads, in our
311 model, to a lower effectiveness than predicted by theory and adds another element of uncertainty to
312 predicting how AOA would work if actually deployed.

313 General surface ocean acidification can be detected in the runs of ensemble C until the year when
314 AOA is initiated (figure S9 in supplementary material). Thereafter total alkalinity accumulates until
315 year 2099 with regional TA reaching concentrations about 200 ~ 500 mmol/m³ higher than the initial
316 values. Comparisons between this study and other AOA studies that included regional applications,
317 such as Ilyina *et al* (2013a), are difficult because those studies were designed to investigate AOA as a
318 means for global CO₂ mitigation, and thus even when AOA was applied regionally it was in still
319 relatively large areas that have a high potential for increasing the uptake of atmospheric CO₂. An
320 implementation of AOA on a regional scale of less than 10 geographical degrees across for only a
321 short time of less than 100 years, has only a limited impact on atmospheric CO₂, while a global
322 implementation of AOA (Keller *et al* 2014), in particular when applied for centuries to millennia
323 (Ilyina *et al* 2013a), can significantly impact atmospheric CO₂ and the global carbon cycle. In contrast
324 to results of the global AOA studies, only a relatively low carbon sequestration and storage potential,
325 with less than a 20 ppm atmospheric CO₂ reduction, is achieved in our regional AOA simulations. In
326 Keller *et al* (2014) a global implementation of lime-based AOA is deployed from year 2020 to year
327 2100 leads to an atmospheric CO₂ decrease about 166 ppm, while Ilyina *et al* (2013a) observe a CO₂
328 drawdown of up to 450 ppm in their global and “Atlantic+Pacific” AOA implementation scenarios.
329 Our results imply that from the regions we selected, regional and decadal- to centennial-scale AOA
330 would not be an appropriate means for significant climate remediation.

331 Differences between regional and global AOA also affect the local seawater chemistry after a
332 termination of AOA. If regional AOA is terminated abruptly, regional seawater pCO₂, aragonite Ω and
333 pH rapidly return to the levels found in the control run (figure 3) on an annual timescale. This is
334 different from the findings of large-scale AOA simulations where such a termination effect is not
335 observed (Ilyina *et al* 2013a, Keller *et al* 2014). In the case of regional AOA, lime and the dissolution
336 products are dispersed rapidly and diluted by seawater from outside the deployment area. Such a rapid
337 change in regional ocean chemistry, which is faster than in any climate change scenario, could
338 potentially put substantial stress on regional ecosystems. Thus, if regional AOA was done without

339 reducing atmospheric CO₂, the process of adding lime would potentially have to continue for very
340 long times or be phased out carefully to avoid risks to coral reef ecosystems.

341 A practical consideration is how much our optimal AOA applications would cost. For CaO-based
342 AOA (Ca(OH)₂ is hydrated CaO) cost estimates provided by Renforth *et al* (2013) indicate that every
343 ton of CO₂ taken up by the ocean as a result of AOA costs approximately \$72-159 (US dollars). These
344 estimates include the extraction, calcination, hydration, and surface ocean dispersion costs associated
345 with AOA and are likely higher than AOA in our study would be since the transportation costs were
346 based on covering the entire global ocean. In our optimal simulation, the cumulative amount of
347 atmospheric CO₂ that is sequestered by AOA in the year 2099 is 56.32 Gt CO₂ in the GB, 119.28 Gt
348 CO₂ in the CS and 129.84 Gt CO₂ in the SC. Based on Renforth *et al* (2013), AOA would cost around
349 US\$ 51-112 billion for the GB per year (if we assume an even sharing of costs over the 80 year
350 periods of our AOA simulations), US\$ 107-237 billion for the CS per year, and US\$ 117-258 billion
351 for the SC per year. Among all three studied regions, GB has the largest number of coral reef locations
352 and the AOA costs for it are the lowest from our study. The Gross Domestic Product (GDP) for
353 Australia in the year 2014 was US\$ 1.45 trillion. According to our model results, Australia could keep
354 the GB region from crossing our chosen guardrails by spending 3.5% ~ 7.7% of its GDP for coral reef
355 protection. Admittedly, this is a huge investment compared with the estimated benefits (5~7 billion
356 US\$ per year) related to coral reefs (GBRMPA (2013)).

357 Ocean acidification is only one of the stressors that corals reefs face in the future. Our study has
358 not addressed other problems such as overfishing (Loh *et al* 2015) or thermal stress (Goreau and
359 Hayes 1994). Coral reef bleaching, caused by thermal stress, is one of the most lethal and enduring
360 threats to coral reefs. For example, in the Great Barrier Reef, between 11 to 83% of coral colonies
361 were affected by large-scale bleaching due to unusually high temperatures during 1998, an El-Niño
362 year, with the mortality rate varying between 1% and 16% (Marshall and Baird 2000). The GB coral
363 coverage declined by around 50% between 1985 to 2012, with 10% of the total loss attributed to coral
364 bleaching (De'ath *et al* 2012). In the Caribbean Sea (CS), thermal stress in the year 2005 exceeded
365 observed levels in the previous 20 years causing over 80% of corals to bleach and resulting in a 40%
366 population loss (Eakin *et al* 2010). In the South China Sea (SC), massive coral bleaching in 1997 and
367 1998 affected 40% of coral colonies, but many of them recovered within a year (Waheed *et al* 2015).
368 Model simulations have suggested that coral bleaching incidents will increase with global warming,
369 and the threat will become more severe in the future if CO₂ emissions remain high and significant
370 warming occurs (Frieler *et al* 2012, Caldeira 2013). According to Teneva *et al* (2012)'s model
371 simulations, our three study areas are in coral bleaching hot spots (Goreau and Hayes 1994) with a
372 middle to high likelihood of experiencing bleaching. Donner (2009) predicted that the 10 years' mean
373 SST during 2090 to 2099 in our studied regions are 3.2°C (SC), 3.3°C (GB) and 3.4°C (CS) higher

374 than those during 1980 to 2000 under a business-as-usual high CO₂ emission scenario. Given such
375 predictions, the question arises as to whether or not regional AOA would be sufficient if CO₂
376 emissions remain high, e.g., warming might harm coral reefs long before acidification becomes a
377 significant threat. There have been proposals to use cloud brightening (Latham *et al* 2013) to cool
378 down surface temperatures to prevent coral bleaching and it is possible that other solar radiation
379 management (SRM) methods may be envisaged in a similar manner. If SRM were seriously
380 considered for this purpose when atmospheric CO₂ levels are high, AOA would be worth considering
381 as well.

382

383 **5. Conclusions**

384 Our results show that with simulated AOA, regional surface aragonite Ω and pCO₂ could be
385 prevented from crossing the acidification thresholds that we set (pCO₂ < 500 μ atm, Ω > 3). In this
386 respect, marine biota could benefit from AOA. To successfully protect corals and associated marine
387 biota from OA within all three regions examined in our study, one would need to deploy about 356 Gt
388 lime over next 80 years, with estimated implementation costs between 275 and 607 billion US dollars
389 annually. This can possibly “buy sometime” before ocean acidification induces physiological stress
390 and ecological shifts. We have also shown that the carbon sequestration potential of regional AOA is
391 small, with regional differences in its effectiveness and sensitivities. Due to rapid exchange with
392 untreated waters from outside the regions, a termination effect would have to be taken into account
393 should deployment of regional AOA be considered in reality. This research shows that AOA has the
394 potential to mitigate regional ocean acidification for the purpose of protecting tropical coral reef
395 ecosystems. Details about environmental side effects will have to be explored with higher resolution
396 models and dedicated lab and possibly field experiments. From a climate change perspective the best
397 solution would obviously be to stop emitting CO₂ and thereby prevent warming and ocean
398 acidification from occurring and affecting coral reef ecosystems in the first place. Since this is unlikely
399 to happen in the near future, it is worth investigating climate engineering methods such as AOA, since
400 they might be able to provide an alternative or complementary means of protection.

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405 **Acknowledgments**

406 This is a contribution to the SPP 1689 ‘Climate Engineering - risks, challenges, opportunities?’
407 funded by the Deutsche Forschungsgemeinschaft (DFG). Additional funding was provided by the

408 BMBF BIOACID Program (FKZ 03F0608A) to W.K. All authors declare that they have no potential
409 conflicts of interests.

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