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# Significant long-term increase of fossil fuel CO<sub>2</sub> uptake from reduced marine calcification

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## Abstract

Analysis of available plankton manipulation experiments demonstrates a previously unrecognized wide range of sensitivities of biogenic calcification to simulated anthropogenic acidification of the ocean, with the “lab rat” of planktic calcifiers, *Emiliana huxleyi* not representative of calcification generally. We assess the implications of the experimental uncertainty in plankton calcification response by creating an ensemble of realizations of an Earth system model that encapsulates a comparable range of uncertainty in calcification response. We predict a substantial future reduction in marine carbonate production, with ocean CO<sub>2</sub> sequestration across the model ensemble enhanced by between 62 and 199 PgC by the year 3000, equivalent to a reduction in the atmospheric fossil fuel CO<sub>2</sub> burden at that time of up to 13%. Concurrent changes in ocean circulation and surface temperatures contribute about one third to the overall importance of reduced plankton calcification.

## 1 Introduction

Open ocean dwelling calcifying plankton, such as coccolithophorids and foraminifera produce shells by precipitating the mineral calcium carbonate (CaCO<sub>3</sub>). The continued dissolution of fossil fuel CO<sub>2</sub> in the surface waters of the ocean and associated decrease in ambient carbonate ion (CO<sub>3</sub><sup>2-</sup>) concentration (Orr et al., 2005) and pH (termed ocean acidification; Caldeira and Wickett, 2003) will adversely affect the ability of these species of plankton to precipitate CaCO<sub>3</sub> (e.g., Bijma et al., 1999; Riebesell et al., 2000; Zondervan et al., 2001). A reduction in marine carbonate production will, in turn, increase the capacity of the surface ocean to absorb CO<sub>2</sub> (Barker et al., 2003; Zondervan et al., 2001). Understanding the strength of this “CO<sub>2</sub>-calcification” feedback (the acceleration of CO<sub>2</sub> uptake by the ocean due to lower calcification rates) and its time-scale of impact is essential to accurate prediction of the fate of CO<sub>2</sub> emitted to the atmosphere and the extent of future climate change. We address this by collating

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available information regarding the calcification responses of different marine organisms to changing  $\text{CO}_3^{2-}$ , and directly quantifying the implications of our findings for the future sequestration of fossil fuel  $\text{CO}_2$  using an Earth system model.

## 2 Synthesis of experimental calcifier response

5 The response of a number of different calcifying marine organisms to changes in ambient carbonate chemistry has been investigated to date (summarized in Table 1). However, the experimental setups and chosen manipulations of the carbonate system differ, hindering simple comparison. We address this by calculating the relative change in calcium carbonate production (calcification rate) as a function of a common variable. We  
10 adopt the widely-used description of the precipitation of carbonate minerals (Zhong and Mucci, 1993) as occurring at a rate equal to;

$$k \times (\Omega - 1)^n \quad (1)$$

where  $\Omega$  is the saturation state (or solubility ratio) of the aqueous environment, defined as  $[\text{Ca}^{2+}] \times [\text{CO}_3^{2-}] / K_{sp}$  (with  $K_{sp}$  a solubility constant), and  $k$  is an overall scaling  
15 constant.

For each set of experimental manipulations detailed in Table 1,  $\Omega$  of calcite is calculated from mean dissolved inorganic carbon and alkalinity concentrations, salinity, and temperature measured over the course of each experiment, and using the dissociation constants of Mehrbach et al. (1973), as refitted by Dickson and Millero (1987). For  
20 *Emiliania huxleyi* (in vitro) (Zondervan et al., 2001) (#1) we combine the two different light-dark cycle data-sets, whereas there is sufficient experimental data to fit the results of “high light” and “complete darkness” incubations of the foraminifera *Orbulina universa* (in vitro) (Bijma et al., 1999) (#2). For the *E. huxleyi* mesocosm experiments (Delille et al., 2005). (#3), we fit all the individual replicates across the three treatments.  
25 Finally, for the ship-board incubations of phytoplankton assemblages (Riebesell et al.,

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2000) (#5), we first normalize the calcification rate data to a value of 1.0 at the common  $\text{CO}_2$  concentration of  $10 \mu\text{mol l}^{-1}$ , before fitting the results of all 5 experiments together.

The fitting procedure is carried out assuming Eq. (1), and constraining the curve to go through the origin (i.e., zero calcification at  $\Omega=1$ ). 100 iterations of a Levenberg-Marquardt algorithm are used to minimize the cost function, and we report the standard error associated with the estimated fitted value of  $\eta$ . The value of the parameter  $\eta$  is crucial here, since it represents the degree of non-linearity of response of calcification rate to a change in ambient carbonate ion ( $\text{CO}_3^{2-}$ ) concentration (and thus  $\Omega_p$ ). A high value of  $\eta$  implies that even a modest anthropogenically-driven reduction in ocean  $\text{CO}_3^{2-}$  (and  $p\text{H}$ ) might drive a substantial decrease in  $\text{CaCO}_3$  production, significantly enhancing fossil fuel  $\text{CO}_2$  uptake by the ocean.

### 3 Future impacts of reduced marine calcification on fossil fuel $\text{CO}_2$ uptake

The range in sensitivities exhibited by calcifying plankton coupled with uncertainty in which plankton groups dominate past, present, and future marine carbonate production, means that no single estimate of the strength of  $\text{CO}_2$ -calcification feedback can currently be made with any confidence. We approach this problem by means of an ensemble Kalman filter (EnKF) calibration of ocean carbon cycling in the GENIE-1 Earth system model (comprising; 3-D ocean circulation model, 2-D energy-moisture balance model (EMBM) of the atmosphere, dynamic-thermodynamic sea-ice model, and representation of atmosphere-ocean-sediment carbon cycling) (Ridgwell et al., 2006<sup>1</sup>). In this, we utilized observed 3-D distributions of alkalinity (Key et al., 2004) and phosphate (Conkright et al., 2002) in the ocean, together with the 2-D distribution of  $\text{CaCO}_3$  content in surface sediments (Archer, 1996) to constrain model parameters controlling the marine carbon cycle. We employ Eq. (1) in calculating  $\text{CaCO}_3$  production (in relation

<sup>1</sup>Ridgwell, A. and Hargreaves, J. C.: An efficient Earth System Model of Global biogeochemical cycling: Regulation of atmospheric  $\text{CO}_2$  by deep-sea sediments, submitted, [http://www.seao2.org/pubs/ridgwell\\_hargreaves\\_manuscript.pdf](http://www.seao2.org/pubs/ridgwell_hargreaves_manuscript.pdf), 2006b.

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to particulate organic carbon (POC) export), and calibrate both  $k$  and  $\eta$  in the EnKF (Ridgwell et al., 2006a; Ridgwell et al., 2006b<sup>1</sup>). Note that while POC production is affected by changes in climate (such as stratification), for simplicity, we do not modify POC production according to changes in  $pH$  (Zondervan et al., 2001).

Although all ensemble members are tuned tightly to the observations, we obtain a large range of  $\eta$  across the ensemble members (0.19 to 1.55) because the scaling constant  $k$  (Eq. 1) introduces a degree of freedom in fitting modern ocean alkalinity and sedimentary  $\text{CaCO}_3$  distributions. The values we obtain for  $\eta$  thus strongly reflect our prior assumptions (1 standard deviation of  $\eta$  spanning the range 0–1.5), which are informed by the results of our analysis of experimental manipulations (Table 1). The relationship between  $k$  and  $\eta$  is determined by the EnKF itself, consistent with the ocean and sediment data. Thus, although this means that we cannot deduce the most probable value for  $\eta$  from marine geochemical observations using our model, we can address the sensitivity of the marine carbon cycle to the wide range of uncertainty in  $\eta$  by carrying out experiments using the full 54-member ensemble of model instances (rather than a just single model composed of the parameter value means).

We run the 54 member ensemble under a single illustrative  $\text{CO}_2$  emissions scenario, based on the assumption of all conventional fossil fuel reserves (ca. 4000 PgC) being used up sometime early in the 24th century (Lenton et al., 2006; Ridgwell et al., 2006b<sup>1</sup>) (Fig. 1a), slightly less than that assumed by Caldeira and Wickett (2003). Note that for clarity, we do not include interactions with carbonate preservation in deep-sea sediments in the quantification of  $\text{CO}_2$ -calcification feedback. The ensemble members are run in each of four permutations of feedbacks operating; (i) no  $\text{CO}_2$ -calcification or  $\text{CO}_2$ -climate feedbacks, (ii) just the  $\text{CO}_2$ -climate feedback, (iii) just the  $\text{CO}_2$ -calcification feedback, and (iv) both  $\text{CO}_2$ -calcification and  $\text{CO}_2$ -climate feedbacks. The numerical efficiency of the GENIE-1 model is critical, as this combined sensitivity + feedback analysis represents a total of;  $4 \times 54 \times 3000 = 648\,000$  years of coupled 3-D ocean carbon cycle/climate model simulation, which we achieve in just a few days on a small computing cluster.

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The evolution with time of the strength of the CO<sub>2</sub>-calcification feedback is shown in Fig. 1 for the mean ensemble model, and for the individual members in Fig. 2. We find that historical fossil fuel CO<sub>2</sub> emissions drive an ensemble mean reduction in calcite saturation state ( $\Omega$ ) from 5.2 to 4.5 by the year 1994 (closely paralleling previous predictions of the decline in aragonite saturation; Kleypas et al., 1999). This leads to a 13% decrease in marine carbonate production (Fig. 1d) as a consequence of Eq. (1). The contribution to the 1994 oceanic anthropogenic CO<sub>2</sub> inventory (estimated at 118±19 PgC; Sabine et al., 2004) due to CO<sub>2</sub>-calcification feedback ( $\Delta$ CO<sub>2</sub>) and including climate feedbacks in the model is no more than 3.2 PgC (Fig. 2), even under the end-member case of  $\eta=1.55$ .

The scatter across the ensemble compared to the best fit line (Fig. 2) reflects the influence of factors other than differences in the value of  $\eta$  that affect CO<sub>2</sub> sequestration. For instance, the ensemble exhibits initial (year 1765) global CaCO<sub>3</sub> export of between 1.05 and 1.59 PgC yr<sup>-1</sup>, similar to recent data-based production estimates of 0.8–1.4 PgC yr<sup>-1</sup> (Feely et al., 2005; Jin et al., 2006). Because there is no significant correlation with the value of  $\eta$  across the ensemble ( $R^2\sim 0.03$ ), our model experiment encapsulates a sensitivity test of the strength of CO<sub>2</sub>-calcification feedback to uncertainties in initial (pre-industrial) global carbonate production. The relatively muted scatter ( $R^2>0.94$ ) of the ensemble members about the best fit line (Fig. 2) indicates that CO<sub>2</sub> sequestered due to ocean acidification is relatively insensitive to assumptions regarding initial carbonate production in the open ocean.

Ocean saturation and marine carbonate production decline (Fig. 1d) until around the year 2300 when a peak atmospheric CO<sub>2</sub> concentration of ~1400 ppm is reached in response to emissions. Our model ensemble predicts the additional sequestration of 33–107 PgC due to CO<sub>2</sub>-calcification feedback at this time. Although marine carbonate production starts to recover thereafter,  $\Delta$ CO<sub>2</sub> continues to increase, reaching 62–199 PgC by the next Millennium.

The persistence of the impact of reduced calcification reflects the time needed to establish a new alkalinity (ALK) structure in the ocean (Fig. 3). By the year 3000 a pro-

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found reorganization has occurred, in which the ALK gradients induced by nitrate uptake and remineralization start to dominate over the currently much stronger opposite effect due to the precipitation and dissolution of  $\text{CaCO}_3$ . For instance, ALK becomes almost as high in the deep North Atlantic compared to the North Pacific by year 3000, whereas in the modern ocean there is a very pronounced Atlantic→Pacific gradient.

## 4 Discussion

Our analysis reveals that calcifying organisms studied in vitro exhibit a wide range of sensitivities of calcification to saturation state (Table 1). In contrast, marine ecosystem models invariably assume a single species (*Emiliana huxleyi*) as representative of carbonate production in the open ocean (e.g., Moore et al., 2002; Le Quéré et al., 2005). This is problematic because while *E. huxleyi* forms large blooms over wide areas of the ocean is not thought to be a particularly important calcifying coccolithophorid (Broerse et al., 2000; Ziveri and Thunell, 2000). We find that this species also exhibits the lowest observed calcification sensitivity to a change in  $\text{CO}_2$ . Furthermore, foraminifera such as *Orbulina universa* which appear to be rather more sensitive than *E. huxleyi*, may dominate carbonate export globally (Schiebel, 2002).

We also find substantial differences between the results of different experimental methodologies with the same species – for instance, an approximate tripling of atmospheric  $\text{CO}_2$  concentration drove a 10–19% reduction in carbonate production by *Emiliana huxleyi* when studied in vitro (Riebesell et al., 2000; Zondervan et al., 2001) but 42% under a similar  $\text{CO}_2$  change in a mesocosm bloom experiment (Delille et al., 2005). Furthermore, incubated natural phytoplankton assemblages from the North Pacific have shown decreases in the rate of calcification of up to 83% over this same atmospheric  $\text{CO}_2$  range (Riebesell et al., 2000). Although the assemblage incubation results may also reflect a delay in coccolith production (Delille et al., 2005), we think that the apparent progression observed in carbonate production sensitivity; in vitro → mesocosms → natural assemblages is important. We suggest it reflects either a

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higher calcification susceptibility of un-characterized calcifying plankton species, or an amplification of acidification impacts on carbonate production by ecosystem dynamics, perhaps reflecting a lower fitness of calcifying plankton which would result in fewer individuals in addition to less  $\text{CaCO}_3$  production per individual.

5 The enhancement by of the marine sequestration of fossil fuel  $\text{CO}_2$  by the next Millennium (Fig. 2) as a result of reduced surface ocean calcification represents up to 13% of the atmospheric fossil fuel  $\text{CO}_2$  burden at this time (the anthropogenic excess over the pre-industrial inventory). Thus, while  $\text{CO}_2$ -calcification feedback reduces the peak atmospheric  $\text{CO}_2$  concentration by  $\sim 15\text{--}49$  ppm in our experiment, its importance  
10 relative to atmospheric  $\text{CO}_2$  is rather greater on the millennial-scale. This has implications for the long-term stability of the Greenland and Antarctic ice caps, which models suggest are very sensitive to the magnitude of protracted greenhouse warming (Alley et al., 2005).

Previous modeling studies of the  $\text{CO}_2$ -calcification feedback (Barker et al., 2003;  
15 einze, 2004; Zondervan et al., 2001) have been unable to account for the role of further amplifying feedbacks between  $\text{CO}_2$  and ocean temperature and circulation. For instance, the year 2100 enhancement of marine  $\text{CO}_2$  sequestration due to  $\text{CO}_2$ -calcification feedback in a recent ocean GCM analysis (Heinze, 2004) was 3.5 ppm ( $\sim 7$  PgC) (Fig. 2) – consistent with our model when assuming the same calcification  
20 sensitivity (*Emiliana huxleyi* in vitro) and invariant climate as per Heinze (2004). However, because our Earth system model includes a climate component (Edwards and Marsh, 2005), we are able to take into account the impact of changes in climate. We find that increased stratification and ocean surface temperatures enhance  $\text{CO}_2$  sequestration arising from reduced calcification by up to one third (Fig. 2) (Although initially,  
25 there is little net effect of climate change on  $\text{CO}_2$ -calcification feedback, and the year 2100 estimate is almost no different.)

It should be noted that we have not taken into account the possibility that reduced carbonate production could affect the transport of organic matter into the deep ocean (Ridgwell, 2003) – the ballast hypothesis (Armstrong et al., 2002). The consequence

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of such a mechanistic dependence of organic carbon transport on the carbonate flux would be a reduction in the strength of CO<sub>2</sub>-calcification feedback (Barker et al., 2003; Heinze, 2004). However, alternative interpretations of sediment trap correlations do not require the efficiency of the organic carbon pump to be dependent on the CaCO<sub>3</sub> flux (Francois et al., 2002; Passow and De La Rocha, 2006). Our model parameterization also implicitly assumes that calcification ceases at  $\Omega=1.0$  (Eq. 1). While for corals, the general trend across a range studies is consistent with our assumption (Langdon and Atkinson, 2005), organisms exerting a much stronger degree of control over the calcification process, particularly foraminifera, need not be so constrained. Conversely, simultaneous decline in coral calcification (Kleypas et al., 1999) as well as physiological (rather than climatic) CO<sub>2</sub> feedbacks on POC production (an effect which is substantial in *E. huxleyi* and *G. oceanica*; Zondervan et al., 2001) would act to increase the sequestration of fossil fuel CO<sub>2</sub>.

## 5 Conclusions

Our analysis highlights the importance of the prevailing uncertainties regarding the controls on marine carbonate production and calcifier response to anthropogenic ocean acidification. We conclude from the experimental data currently available that the response of carbonate production to a change in environmental chemistry as measured in vitro is not a good indication of the ecosystem (or global) level response. We also find that the behavior of the coccolithophorid *Emiliana huxleyi* to increasing CO<sub>2</sub> is rather small compared to other coccolithophores species, foraminifera and corals. Ecosystem models that adopt this species as their “functional type” for carbonate production will underestimate the importance of CO<sub>2</sub>-calcification feedback on future climate change.

There is thus a strong argument for further mesocosm experiments and higher level manipulations of more complete ecosystems as well as better quantification of which calcifying species control CaCO<sub>3</sub> production globally and the spatial distribution and (environmental and ecological) controls on this production. However, when faced with

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current uncertainties, the utility of computationally efficient Earth System Models is clear.

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**Table 1.** Analysis of experimental calcification rate manipulations.

#	Species	Experimental details	$\Omega$ (calcite)	Estimated $\eta$ <sup>1</sup>	reference
1	<i>Emiliana huxleyi</i>	in vitro	2.0–4.8	0.20±0.09	Zondervan et al. (2001)
2	<i>Orbulina universa</i>	in vitro (high light)	1.0–4.1	0.32±0.08	Bijma et al. (1999)
2	<i>Orbulina universa</i>	in vitro (darkness)	1.7–4.1	0.49±0.23	Bijma et al. (1999)
3	<i>Emiliana huxleyi</i>	mesocosm bloom	2.0–5.3	0.32±0.20	Delille et al. (2005)
4	<i>Gephyrocapsa oceanica</i>	in vitro	1.7–4.3	0.53±0.31	Zondervan et al. (2001)
5	Mixed assemblage	Ship-board incubation	1.7–4.4	0.75±0.10	Riebesell et al. (2000)
6	Corals	(various)	1–6 <sup>2</sup>	~ 1	Langdon and Atkinson (2005)
7	None (abiotic)	in vitro		1.9–2.8	Zhong and Mucci (1993)

<sup>1</sup> mean value and standard error

<sup>2</sup> saturation state of aragonite

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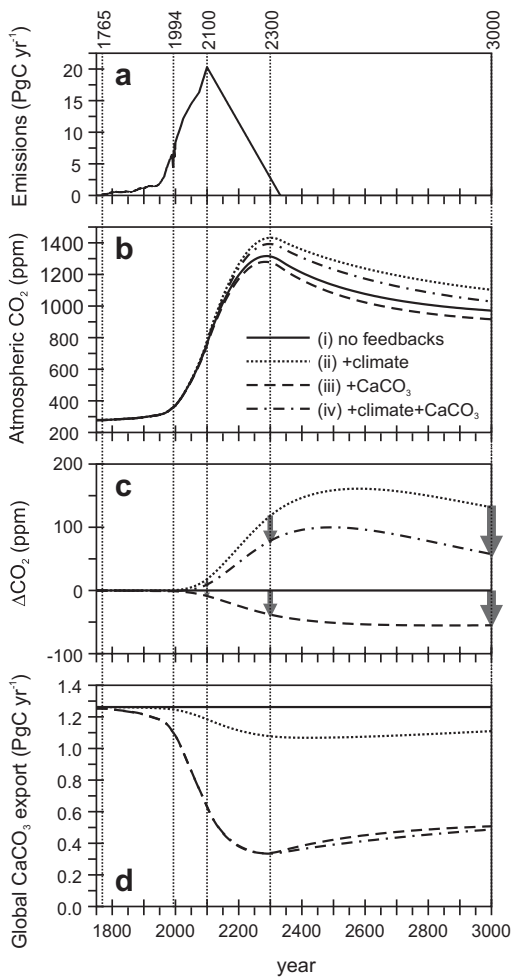


Fig. 1.

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**Fig. 1.** Atmospheric CO<sub>2</sub> and marine carbonate production responses to assumed fossil fuel emissions.

A single model instance employing the ensemble mean parameter set <sup>1</sup> rather than all individual members of the ensemble is used here to illustrate the impact of CO<sub>2</sub>-calcification feedback. **(a)** Time-history of the rate of CO<sub>2</sub> emissions to the atmosphere prescribed in the model (see refs. Lenton et al., 2006, Ridgwell et al., 2006b<sup>1</sup>). **(b)** Evolution of atmospheric CO<sub>2</sub> in experiments with; (i) neither climate nor calcification changes (solid line), (ii) “CO<sub>2</sub>-climate” feedbacks operate (dotted line), (iii) calcification responds to atmospheric CO<sub>2</sub> (i.e., “CO<sub>2</sub>-calcification” feedback) (dashed line), and (iv) both CO<sub>2</sub>-calcification and CO<sub>2</sub>-climate feedbacks operate (dot-dash line). **(c)** The impact on the evolution of atmospheric CO<sub>2</sub> of the two feedbacks is shown, both individually, and combined (and compared to the baseline (i.e., no feedbacks) scenario). The grey arrows indicate the strength of CO<sub>2</sub>-calcification feedback ( $\Delta\text{CO}_2$ ) at different time-slices (years; 1994, 2100, 2300, 3000) marked with vertical dotted lines. **(d)** The response of global carbonate export under the different permutations of feedback. In the absence of any response of calcification to changing carbonate saturation, CaCO<sub>3</sub> export declines in response to fossil fuel emissions as a result of changes in ocean circulation (stratification) when feedbacks between CO<sub>2</sub> and climate are allowed to operate. (Legend as per 1b.)

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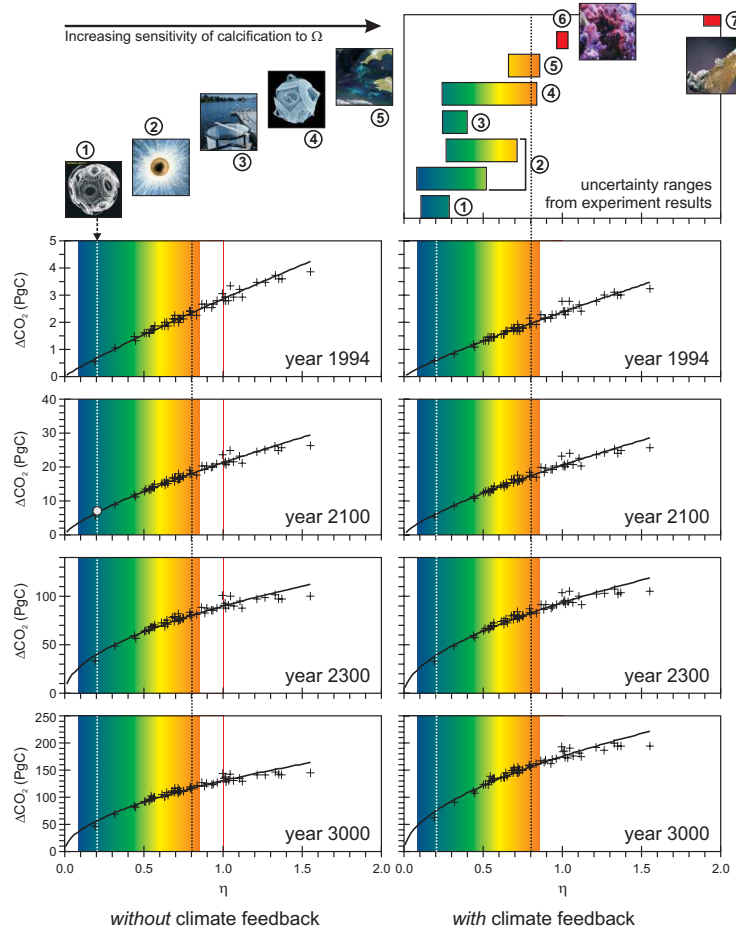


Fig. 2.

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**Fig. 2.** Response of the 54-member ensemble of realizations of the GENIE-1 Earth system model to CO<sub>2</sub> emissions.

The strength of the CO<sub>2</sub>-calcification feedback (see Fig. 1) is plotted against the calcification rate sensitivity ( $\eta$ ) (Eq. 1) for each of 4 time-slices. Crosses represent individual ensemble members and the solid line is a power law fit ( $R^2 > 0.94$  in each case). Results are shown for the strength of the CO<sub>2</sub>-calcification feedback both with (RH side panels) and without (LH side) further feedbacks with climate. The filled grey circle represents the year 2100 prediction of the ocean GCM of Henize (2004), which assumed the calcification sensitivity of *Emiliana huxleyi* (Zondervan et al., 2001) (indicated by the vertical dotted white line) and no feedbacks with climate (and calculated at a comparable atmospheric CO<sub>2</sub> concentration). The values of  $\eta$  corresponding to the characteristics of different calcification systems are highlighted at the top, with mean  $\eta$  value and uncertainty limits as given in Table 1; #1, the coccolithophorid *Emiliana huxleyi* (in vitro) (Zondervan et al., 2001), #2, the foraminifera *Orbulina universa* (in vitro) (Bijma et al., 1999), #3, *E. huxleyi* mesocosms (Delille et al., 2005), #4, the coccolithophorid *Gephyrocapsa oceanica* (in vitro) (Zondervan et al., 2001), and #5, ship-board incubations of phytoplankton assemblages (Riebesell et al., 2000). Also shown for comparison are; #6, corals (as summarized by Langdon and Atkinson, 2005), and #7, “abiotic” precipitation (i.e., occurring in the absence of any biological intervention) (Zhong and Mucci, 1993). The ensemble mean value ( $\eta = 0.81$ ) is indicated by a vertical dotted black line.

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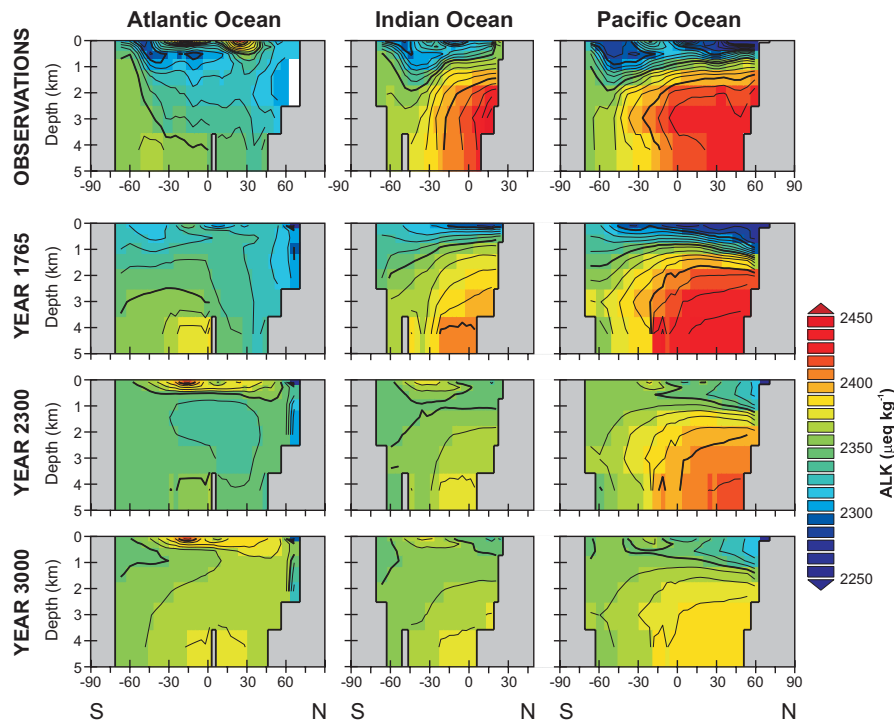
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**Fig. 3.** Reorganization of the ocean alkalinity structure.

The zonal mean distribution of alkalinity (ALK) undergoes a large-scale reorganization in response to the reduction in marine carbonate production. Shown compared to observations (Key et al., 2004) (top panel) are the predictions of the ensemble mean model ( $\eta = 0.81$ ) for the years; 1765, 2300, and 3000.

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