General comments
I was very pleased to see that the authors incorporated many of the changes suggested by the
three reviewers, most notably the sensitivity analysis. This has substantially improved the
quality of the manuscript, and has moved away the paper from just being a comparison with the
output from CMIP5 Earth System Models. The manuscript also clearly benefits from the
improved model description, separating the Results and Discussion sections, and improved
design of the tables and figures.

Personally, I disagree with the authors that their original Figure 7 didn’t add anything to the
manuscript, but can live with them taking it out as its original discussion contained some flaws.
What still requires improvement is the description of the sensitivity analysis. Although the
manuscript is definitely better structured compared to the first version and many typos have
been removed, it is still left with some ambiguous statements and points that need clarification,
mostly in the newly added parts.

*Due to the great comments from the reviewers the manuscript is significantly improved.

Specific comments
- Lines 26-28: do you mean ‘sensitive’ or rather ‘most sensitive’? The current statement implies
that pH and Ωar are not sensitive at all to other parameters; is this true? Also, add ‘Of the
parameters tested’ at the beginning of the sentence; the statement now implies that all
parameters were tested.

*We mean ‘most sensitive’. Thank you for picking this up.

- Line 53: where does this 10% come from? Provide a reference. For example, Sarmiento and
Gruber (2006) mention that currently ca. 1 out of 20 molecules of CO2 that dissolve in seawater
(i.e. 5%) will remain in this form.
*calculated from refit by Mehrback refit from Dickson and Millero, 1987

- Line 92: what are ‘non-CO2s’? Please explain
* “…which takes in emissions of CO2, non-CO2s, such as CH4, N2O and halocarbons, and aerosols,..”

- Lines 94-96: If this is a new component of Hector compared to Hartin et al. (2015), then this
can be detailed a bit more, for example by explaining this new algorithm in Appendix A.
*There are no new comments between Hector v1.0 and Hector v1.1. between the versions we
updated the API for linking with external models, updated the backend R scripts, and some
minor bug fixes.

- Line 116 (equation 3): shouldn’t the left hand side of this equation be F(i→j)? If not, how is
this equation related to equation 2?
*yes, this has been corrected.

\[- Lines 126-129: The authors do mention now that all carbon in the ocean compartment is in
the form of DIC and ratify this for DOC, but not for POC. Please add references showing how of
ocean’s carbon is in the form of POC and why it can be neglected in Hector.
We added a small sentence addressing POC. “Dissolved organic matter is less than 2% of the total inorganic carbon pool, of which a small fraction is dissolved organic carbon (Hansell and Carlson, 2001) and particulate organic carbon less than 1% of the total carbon pool (Eglinton and Repeta, 2004).”

- Lines 146: why not state here that \( k = 0.585 \text{Sc}^{-1/2} U_{10}^{-2} \) and later simply define \( T_r \) as the product of \( \alpha \) and \( k \)?
  *while this also is correct, we decided that the original description corresponds to the model code better, making it easier to track the flow of commands.

- Lines 169-174: I really like the inclusion of the sensitivity analysis and I’m perfectly fine with not testing all parameters in it. This description is, however, not detailed enough. What I miss here, however, is (a) which parameters were selected; (b) why you thought a priori that these parameters were important for the marine carbonate system; and (c) (this also holds for Table 7) it needs to be made clear where the selected parameters appear in Hector. For salinity and temperature this is quite clear, also because they are given in Table 1, but for e.g. albedo and beta (what does beta even stand for? I see it is defined in line 265, but it should be added to Table 7) this isn’t clear at all.
  *This is a great comment by the reviewer. We have added throughout the text some significant improvements to the description of the parameters. For example, “We selected those parameters directly influencing atmospheric \( \text{CO}_2 \) concentrations (beta and \( Q_{10} \)), parameters involved in the calculation of temperature (albedo and climate sensitivity) and those parameters involved in the uptake of carbon in the surface ocean (ocean surface temperature, salinity, wind stress, and ocean circulation).”

- Lines 192-193: how exactly were the data annually averaged, e.g. via linear interpolation between sample times before averaging?
  *The data was simply annually averaged. We appreciate the reviewer’s thoughtful point on doing a linear interpolation prior to averaging and we will implement this change in the future.

- Lines 193-195: were the settings in CO2SYS chosen such that they are compatible with the equations used in Hector to calculate the carbonate system? Please state e.g. which equilibrium constants were chosen here and whether Si and P data were included in these calculations (as opposed to Hector where they were not included).
  *We updated the text to reflect these changes. “The carbonate parameters not found in Table 3 are computed from temperature, salinity, and the given carbonate parameter pairs using the CO2SYS software (Lewis and Wallace, 1998). The equilibrium constants (K1 and K2 from Mehrbach et al., 1973 refit by Dickson and Millero, 1987) and total phosphorus and silica set to zero where chosen to best match Hector.”

- Line 196: add “and assuming constant TA” as this is a crucial assumption in these calculations
  *If TA was used as an input to CO2sys, than TA was not held constant.

- Line 173 (and section 3.2): a percentage change in pH is not physically meaningful as pH itself is presented on a logarithmic scale. Please give pH changes in absolute values, or give percentage changes of \( H^+ \) concentration.
  *We agree strongly with the reviewer. We have since changed our results to the percent change for \( [H^+] \).
- Lines 211-212: only at high latitudes is this decline between 2100-2300 clearly visible; for the low latitudes both Hector and CMIP5 seem to show a very slight decrease. Rephrase this text. *updated: “Second, after 2100 the high latitude CMIP5 median begins to decline, while Hector rises and stabilizes.”*
- Lines 269-270: what would be the underlying mechanism for these different responses? For example, why is pH in the high latitude surface ocean more sensitive to wind stress than at low latitudes? Please elaborate a bit on this in the discussion. *Since we switched to percent change of H+ the differences, we found that the high latitude was more sensitive to all changes in parameters at 2100 and 2300. This is most likely due to the fact that the high latitude ocean makes up only 15% of the global ocean.*
- Lines 316-318: It is key to note that this upscaling is only valid if we assume a linear response. This should be added here. Moreover, I’m not convinced at all that it is realistic to expect such a linear response, you already see that in the fact that in Table 7 the percentage changes for +10% and -10% are not mirrored. I’d therefore suggest dropping this statement. *the reviewer raises a good point and we have deleted this statement.*
- Lines 331-333: I’d stress here that this is the case for the open ocean / on the global scale. Locally, in coastal areas, this may not be true and also cannot be shown with Hector. *agree*
- Appendix B (and section 4): nowhere in the manuscript it is mentioned how total borate is defined. This needs to be added to the Appendix. Is it calculated as a function of salinity? If so, it also makes up part of the system response when changing salinity; this should be mentioned in the Discussion. *equation of borate was added to the Appendix.*
- Table 5: Are the presented rates of change the average rates of change over those periods (i.e. 1850-2100 and 1850-2300) or the rates of change in these specific years (i.e. 2100 and 2300)? Please clarify in the figure caption. The same holds for Table 6; in this case it is specified in the text (lines 197-199) but for clarity it should be added to the caption. *we added an extra column to the table with the year ranges*
- Figure 2: I understand the big jump in CMIP5 mean DIC around 2100, switching from n=15 to n=3, but why is there a jump in mean DIC around ~1870? *There are 4 models (2 versions of each GFDL and HadGEM) that do not start until 1859 and 1860. Therefore, when these models appear there is a jump in the graph.*

Technical corrections
*We thank the reviewer for their careful edits of the manuscript. We have since made all necessary changes to the text.*
- Line 24: change ‘saturations’ to ‘saturation levels (Ωar)’ or ‘saturation states (Ωar)’. This definition is necessary as it comes back later in the abstract
- Lines 27-28: don’t just give Q10 and beta here. If I read this without having read the article, I’d think beta refers to acid-base buffering capacity and Q10 to a temperature coefficient in the ocean. Define the parameters or just describe them.
- Line 47: the superscript in H+ is missing.
Review - “Ocean acidification over the next three centuries using a simple global climate carbon-cycle model: projections and sensitivities.”

General issue:
Throughout the manuscript there is a problem with references. These should be written either in the form “Hartin et al. (2015)” or in the form “(Hartin et al., 2015)”. I have found errors on the following lines: 97, 122, 283, 348, 366, 399, and in addition most refs in Table 1 and all refs in Table 3. There could of course be other mistakes that I missed, so please check this carefully.
*Thank you for noticing this issue. We have carefully looked over all of the in-text citations and made all necessary changes.

Other comments:
*We thank the reviewer for their detailed comments below. We have addressed all of these within the manuscript.

Line 26-27: Beta and Q10 are not defined until line 265-267 (and by the way defined again on line 297-298). I think you should explain these parameters already in the abstract.

Line 47: The “+” in “H+” should be in superscript.

Line 69-73: The RCP and CMIP5 acronyms were already defined in the abstract, maybe that’s sufficient.

Line 156 and 362: The “sp” in “Ksp” should be in subscript.

Line 210: “First, he carbon pools...” – change to “the carbon pools”.

Line 233: “Recent work, suggests” – remove comma.

Line 236: “Twice” instead of “double”.

Line 237: Change “decrease in pH of 7.96” to “decrease in pH to 7.96”.

Line 255 and 290: Change “under saturation” to “undersaturation”?

Line 255-257: According to the text: “Even under a best-case scenario, RCP 2.6, low latitude pH will drop to 7.73 by 2100 and to 7.43 by 2300, with ΩAr saturations remaining well outside of present day values”. However, according to Figure 6, pH in the best-case scenario is around 8.1 and 8.15 in 2100 and 2300 respectively, while the aragonite saturation steadily increases... so what happened here? – By the way, I would change “saturations” on line 256 to “saturation levels”.
*Thank you for this. We have no idea where those numbers in the text came from but they have been updated accordingly.

Line 288: Pelejero et al. (2010) is not in the reference list.

Line 320: The Section number should be 5 and not 6.

Line 323: “study, calculates” – remove comma.

Line 328: “archive, includes” – remove comma.

Line 331-332: “Overall, we find that parameters directly involved changes in atmospheric [CO2] have the most impact” – Strange sentence, I think a word is missing somewhere.

Line 369: Rongudai or Tongudai?

Line 376-379: I would change this section to: “C. Hartin designed and carried out the experiments. C. Hartin, B. Bond-Lamberty, and P. Patel developed the model code. A. Mundra processed data and
designed and prepared figures. C. Hartin prepared the manuscript with contributions from all co-authors.”

Line 386: According to the reference list, the Dore et al. paper is from 2009.
Line 414: Solid/dashed arrows: all arrows in this figure appear to be solid in my copy of the manuscript – maybe use different colors instead? * that was a typo, the arrows are curved and not dashed.
Review of: “Projections of ocean acidification over the next three centuries using a simple global climate carbon-cycle model” by C.A. Hartin et al.

General comments:
The paper presents a fast and competent model tool for future projections of the carbonate system. This is something I think is needed as complement to the more complex, computationally expensive earth system models. The study now includes a sensitivity study and the manuscript is improved by it. The paper is interesting, and presents a promising concept. It still needs some smaller corrections, but nothing major.
*We thank the reviewer for their detailed comments. We have made all necessary changes to the manuscript.

Specific comments:
I have no major specific comments at this stage in the review process.

Line-by-line corrections:
Line 27: beta and Q10 has not been previously defined here, please do so.
Line 47: superscript the + sign in H+.
Line 210: “he” should be “the”?
Lines 260-261: Is “set of parameters” and “the input parameters” the same parameters? Clarify in the text and please refer the reader to a table (table 7?) were they are listed or list them in the text. As of now, it is too hard to follow.
Lines 263-263: I don’t understand the sentence. Are you comparing the range of omega and ph to atmospheric temperature range in your own model or another model or … something else? Clarify.
Lines 267-268: I think it should be mentioned that also surface ocean temperature (table 7) has a rather large impact on omega. The effect of a 10% change in this parameter gives as large changes in omega as the Q10 change.
*We did add a few words about temperature as well to the aragonite sensitivities.
Line 298: “within the land” sounds weird to me, re-write to “on land” or something?
Line 303: The end of the sentence needs to be re-written.
Lines 332-333: Has this been found anywhere else, or is it new?
Line 388: Homogenize the size of the tables and improve presentation (unless that is done in the final layout anyway)
Line 415: Figure 1 has no dashed arrows that I can see.
Ocean acidification over the next three centuries using a simple global climate carbon-cycle model: projections and sensitivities.

C. A. Hartin*, B. Bond-Lamberty, P. Patel and Anupriya Mundra

Pacific Northwest National Laboratory, Joint Global Change Research Institute at the University of Maryland-College Park, 5825 University Research Court #3500, College Park, MD 20740, USA

*Corresponding author: Corinne.hartin@pnnl.gov

ABSTRACT

Continued oceanic uptake of anthropogenic CO₂ is projected to significantly alter the chemistry of the upper oceans over the next three centuries, with potentially serious consequences for marine ecosystems. Relatively few models have the capability to make projections of ocean acidification, limiting our ability to assess the impacts and probabilities of ocean changes. In this study we examine the ability of Hector v1.1, a reduced-form global model, to project changes in the upper ocean carbonate system over the next three centuries, and quantify the model’s sensitivity to parametric inputs. Hector is run under prescribed emission pathways from the Representative Concentration Pathways (RCPs) and compared to both observations and a suite of Coupled Model Intercomparison (CMIP5) model outputs. Current observations confirm that ocean acidification is already taking place, and CMIP5 models project significant changes occurring to 2300. Hector is consistent with the observational record within both the high (>55°) and low latitude oceans (<55°). The model projects low latitude surface ocean pH to decrease from preindustrial levels of 8.17 to 7.77 in 2100, and to 7.50 in 2300; aragonite saturations levels decrease from 4.1 units to 2.2 in 2100 and 1.4 in 2300 under RCP 8.5.
These magnitudes and trends of ocean acidification within Hector are largely consistent with the CMIP5 model outputs, although we identify some small biases within Hector’s carbonate system. Of the parameters tested, Modeled changes in [\( \text{pH}^- \)] are most sensitive to those parameters that directly affect atmospheric CO\(_2\) concentrations - (\( \beta \text{ and } Q_{\text{10}} \) (terrestrial respiration temperature response) as well as changes in ocean circulation, while changes in \( \Omega_{\text{Ar}} \) saturation levels are sensitive to changes in ocean salinity and \( Q_{\text{10}} \). We conclude that Hector is a robust tool well-suited for rapid ocean acidification projections, sensitivity analyses, and is capable of emulating both current observations and large scale climate models under multiple emission pathways.
1. INTRODUCTION

Human activities have led to increasing anthropogenic emissions of greenhouse gases to the atmosphere. In the first decade of the 21st century CO₂ emissions from anthropogenic sources and land use changes accounted for ~9 Pg C yr⁻¹, with future emission projections of up to 28 Pg C yr⁻¹ by 2100 under Representative Concentration Pathway 8.5 (RCP 8.5) (Riahi et al., 2011). The world’s oceans have played a critical role in lessening the effects of climate change by absorbing 25-30% of the total anthropogenic carbon emissions since 1750 (Le Quéré et al., 2013; Sabine et al., 2011).

In response to this increasing atmospheric burden of CO₂ and increasing oceanic uptake, the oceans are experiencing both physical and biogeochemical changes: surface and deep water warming, reduced subsurface oxygen, and a reduction in calcium carbonate saturation levels and pH (Doney, 2010). Mean surface ocean pH has already decreased by 0.1 units relative to preindustrial times (Caldeira et al., 2003). If current emission trends continue, ocean acidification will occur at rates and extents not observed over the last few million years (Feely et al., 2004; Feely et al., 2009; Kump et al., 2009; Caldeira et al., 2003). Ocean acidification occurs when atmospheric CO₂ dissolves in seawater (CO₂(aq)), forming carbonic acid (H₂CO₃) which dissociates into carbonate (CO₃²⁻) and bicarbonate (HCO₃⁻), and releasing protons (H⁺). The net effect of adding CO₂ to the system increases the concentrations of [H₂CO₃], [HCO₃⁻], and [H⁺], while decreasing [CO₃²⁻] concentrations and lowering the pH. The sum of [HCO₃⁻], [CO₃²⁻] and [CO₂⁺], where [CO₂⁺] = [CO₂(aq)] + [H₂CO₃] represents the total inorganic carbon or dissolved inorganic (DIC) of the system. As CO₂(aq) continues to increase in the ocean it reacts with CO₃²⁻, forming HCO₃⁻, decreasing the fraction of CO₂ that can be readily absorbed by the oceans.

Therefore, because of this buffering capacity of the ocean to buffer chemical changes, a doubling of atmospheric [CO₂] will not correspond to a doubling of [CO₂⁺] but instead will result in an increase on the order of 10% (Dickson and Millero, 1987). Due to both these chemical and physical changes (e.g., warming and stratification), the oceans may become less efficient in the uptake of anthropogenic CO₂ as...
the climate continues to change (Sarmiento and Le Quéré, 1996; Matear and Hirst, 1999; Joos et al., 1999; Le Quéré et al., 2010).

Numerous experiments and observations indicate that ocean acidification will have significant effects on calcifying marine organisms. For example, the rate of coral reef building may decrease, calcification rates of planktonic coccolithophores and foraminifera may be suppressed, and significant changes in trophic level interactions and ecosystems may occur (Cooley and Doney, 2009; Silverman et al., 2009; Fabry et al., 2008; Riebesell et al., 2000). Some coral reefs are believed to already be eroding for parts of the year due to changes in ocean acidification (Yates and Halley, 2006; Albright et al., 2013).

Global surface pH is projected to drop by up to 0.33 units (Gehlen et al., 2014; Orr et al., 2005) and all existing coral reefs will be surrounded by conditions well outside of preindustrial values and even today’s saturation levels (Ricke et al., 2013) under the RCP8.5 scenario.

These model projections of ocean acidification come primarily from Earth System Models (ESMs) that integrate the interactions of atmosphere, ocean, land, ice and biosphere to estimate the present and future state of the climate. ESMs are computationally expensive and typically run using stylized experiments or a few Representative Concentration Pathways (RCPs) (greenhouse gas concentration trajectories used in the Intergovernmental Panel on Climate Change 5th Assessment Report (IPCC, 2013)). This generally limits ESM-based analyses to those scenarios. The occurrence of ocean warming and acidification is consistent across the Coupled Model Intercomparison Project (CMIP5) ESMs, but their rates and magnitudes are strongly dependent upon the scenario (Bopp et al., 2013).

An alternative to ESMs are reduced-form models, relatively simple and small models that can be powerful tools due to their simple input requirements, computational efficiency, tractability, and thus ability to run multiple simulations under arbitrary future emission pathways. This allows for the quantification of arbitrary climate change scenarios, emulation of larger ESMs, as well as in-depth
parameter sensitivity studies and uncertainty analyses (Senior and Mitchell, 2000; Ricciuto et al., 2008; Irvine et al., 2012).

Our goal of this study is to quantify how well Hector, a reduced-form model that explicitly treats surface ocean chemistry, emulates the marine carbonate system of both observations and the CMIP5 archive and explore the parametric sensitivities to Hector’s ocean outputs. The remainder of the paper is organized as follows; section 2, a detailed description of Hector’s ocean component, the data sources and simulations run, section 3, results of the model comparison and sensitivity experiments and lastly, section 4, a discussion of the results.

2.0 Model Description – Hector

Hector is open-source and available at https://github.com/JGCRI/hector. The repository includes all model code needed to compile and run the model, as well as all input files and R scripts to process its output. Hector is a reduced form climate carbon-cycle model, which takes in emissions of CO₂, non-CO₂s (e.g., CH₄, N₂O and halocarbons, and aerosols), converts emissions to concentrations where needed, calculates the global radiative forcing, and then global mean temperature change. Hector contains a well-mixed global atmosphere, a land component consisting of vegetation, detritus, and soil, and an ocean component. In this study we use Hector v1.1, with an updated ocean temperature algorithm to better match the CMIP5 mean. For a detailed description of the land and atmospheric components of Hector, please refer to Appendix A and Hartin et al. (2015).

2.1 Ocean Component

Hector’s ocean component is based on work by Lenton (2000), Knox and McElroy (1984) and Sarmiento and Toggweiler (1984). It consists of four boxes: two surface boxes (high and low latitude), one intermediate, and one deep box. The cold high latitude surface box makes up 15% of the ocean surface area, representing the subpolar gyres (>55°), while the warm low latitude surface box (<55°)
makes up 85% of the ocean surface area. The temperatures of the surface boxes are linearly related to the global atmospheric temperature change, and are initialized at 2°C and 22°C for the high and low latitude boxes respectively. This temperature gradient sets up a flux of carbon into the cold high latitude box and a flux out of the warm low latitude box. The ocean-atmosphere flux of carbon is the sum of all the surface fluxes ($F_i$, n=2).

$$F_O(t) = \sum_{i=1}^{n} F_i(t)$$  

Once carbon enters the high latitude surface box it is circulated between the boxes via advection and water mass exchange, simulating a simple thermohaline circulation. In this version of Hector we do not explicitly model diffusion; simple box-diffusion models and “HILDA” (e.g., Siegenthaler and Joos, 1992) type models are typically in good agreement with observations but are more computationally demanding than a simple box model (Lenton, 2000). The change in carbon of any ocean box $i$ is given by the fluxes in and out with $F_{atm\rightarrow i}$ as the atmospheric carbon flux of the two surface boxes:

$$\frac{dC_i}{dt} = \sum_{j=1}^{in} F_{j\rightarrow i} - \sum_{j=1}^{out} F_{i\rightarrow j} + F_{atm\rightarrow i}$$  

The flux of carbon between the boxes is related to the transport ($T_{i\rightarrow j}, m^3 s^{-1}$) between $i$ and $j$, the volume of $i$ ($V_i, m^3$), and the total carbon in $i$ (including any air-sea fluxes) ($C_i, Pg C$):

$$\frac{dC_{i\rightarrow j}}{dt} = \frac{T_{i\rightarrow j} * C_i(t)}{V_i}$$

Volume transports are tuned to yield an approximate flow of 100 Pg C from the surface high latitude box to the deep ocean box at steady state, simulating deep water formation.

Hector includes calculates four measurable variables of the carbonate system in seawater: DIC, total alkalinity (TA), $CO_3^{2-}$, $HCO_3^-$; pCO$_2$ and pH, any pair of which can be used to describe the entire carbonate system. DIC and TA are the two carbonate variables used to solve the rest of the carbonate
system. surface ocean pH and pCO$_2$-values. These detailed carbonate chemistry equations are based on numeric programs from Zeebe and Wolf-Gladrow, [2001] (Appendix B). We simplified the equations by neglecting the effects of pressure, since we are only concerned with the surface ocean. Hector is run to equilibrium in a perturbation-free mode, prior to running the historical period, ensuring that it is in steady state (Hartin et al., 2015; Pietsch and Hasenauer, 2006). DIC (µmol kg$^{-1}$) in the surface boxes is a function of the total carbon (Pg C) and the volume of the box. All carbon within the ocean component is assumed to be inorganic carbon. Dissolved organic matter is less than 2% of the total inorganic carbon pool, of which a small fraction is dissolved organic carbon (Hansell and Carlson, 2001) and particulate organic carbon less than 1% of the total carbon pool (Eglinton and Repeta, 2004). Therefore, for simplicity we chose not to include dissolved or particulate organic carbon within Hector.

TA is calculated at the end of model spinup (running to equilibrium in an a historical, perturbation-free mode) and held constant throughout the run going forward in time, resulting in 2311.0 µmol kg$^{-1}$ for the high latitude box and 2435.0 µmol kg$^{-1}$ for the low latitude box. These values are within the range of open ocean observations, of 2250.0 – 2450.0 µmol kg$^{-1}$ (Key et al., 2004; Fry et al., 2015). We assume negligible carbonate precipitation/dissolution and no alkalinity runoff from the land surface to the open ocean. Alkalinity is typically held constant with time, which is a reasonable assumption over several thousand years (Lenton, 2000; Zeebe and Wolf-Gladrow, 2001; Glotter et al., 2014; Archer et al., 2009). On glacial-interglacial time scales alkalinity and the dissolution of CaCO$_3$ sediments is an important factor in controlling atmospheric [CO$_2$] (Sarmiento and Gruber, 2006), and thus on these scales Hector will underestimate the oceanic CO$_2$ uptake.

Hector solves for pCO$_2$, pH (total scale), and [HCO$_3$$^-$_], [CO$_3^{2-}$], and aragonite (Ω$_{ar}$) and calcite saturation states (Ω$_{Ca}$) in both the high and low latitude surface ocean boxes. pCO$_2$ is calculated from the concentration of [CO$_2^{2-}$] and the solubility of CO$_2$ in seawater, based on salinity and temperature.
$\text{CO}_2^*$ is calculated from DIC and the first and second dissociation constants of carbonic acid from Mehrbach et al. (1973), refit by Lueker et al. (2000) (Appendix B).

Carbon fluxes between the atmosphere and ocean are calculated (Takahashi et al., 2009):

$$F = k \alpha * \Delta p_{\text{CO}_2} = Tr * \Delta p_{\text{CO}_2}$$  \hspace{1cm} (4)

where $k$ is the CO$_2$ gas-transfer velocity, $\alpha$ is the solubility of CO$_2$ in seawater ($K_0$, Appendix B), and the $\Delta p_{\text{CO}_2}$ is the difference in pCO$_2$ between the atmosphere and ocean. The product of $k$ and $\alpha$ results in $Tr$, the sea-air gas transfer coefficient, where $Tr (g \text{ C m}^{-2} \text{ month}^{-1} \mu\text{atm}^{-1}) = 0.585 * \alpha * Sc^{-1/2} * U_{10}^2$, 0.585 is a unit conversion factor (from mol liter$^{-1}$ atm$^{-1}$ to g-C m$^{-3}$ µatm$^{-1}$ and from cm h$^{-1}$ to m month$^{-1}$) and $Sc$ is the Schmidt number. The Schmidt number (Appendix B) is calculated from Wanninkhof (1992) based on the temperature of each surface box. The average wind speed ($U_{10}$) of 6.7 m s$^{-1}$ is the same over both surface boxes (Table 1). pH (total scale), $[\text{HCO}_3^-]$, and $[\text{CO}_3^{2-}]$ are calculated using the $[\text{H}^+]$ ion and solved for in a higher order polynomial (Appendix B).

Aragonite and calcite are the two primary carbonate minerals within seawater. The degree of saturation in seawater with respect to aragonite ($\Omega_{\text{Ar}}$) and calcite ($\Omega_{\text{Ca}}$) is calculated from the product of the concentrations of calcium $[\text{Ca}^{2+}]$ and carbonate ions $[\text{CO}_3^{2-}]$, divided by the solubility product ($K_{sp}$).

The $[\text{Ca}^{2+}]$ is based on equations from Riley and Tongudai (1967) at a constant salinity of 34.5. If $\Omega = 1$, the solution is at equilibrium, and if $\Omega > 1$ ($\Omega < 1$) the solution is supersaturated (undersaturated) with respect to the mineral.

$$\Omega = \frac{[\text{Ca}^{2+}] [\text{CO}_3^{2-}]}{K_{sp}}$$  \hspace{1cm} (5)

### 2.2. Simulation and experiments

Hector is run under prescribed emissions from 1850-2300 for all four Representative Concentration Pathways (RCP 2.6, RCP 4.5, RCP 6, RCP 8.5) (Moss et al., 2010; van Vuuren et al., 2007). We compare how well Hector can emulate the carbonate system of the CMIP5 median. Our results
section will mainly focus on RCP8.5, exploring the response of the carbonate system under a high emissions scenario.

We also ran a series of model sensitivity experiments to quantify how influential some of Hector’s parameter inputs are on its outputs (in particular, $[H^+]$ and $\Omega_{ar}$). Such sensitivity analyses are important to document model characteristics, explore model weaknesses, and determine to what degree the model behavior conforms to our existing understanding of the ocean system. We do not sample Hector’s entire parameter space, a computationally demanding exercise, but instead choose a list of the parameters that we expect, *a priori*, to be important in calculating the marine carbonate system. We selected those parameters directly influencing atmospheric CO$_2$ concentrations (beta and $Q_{10}$), parameters involved in the calculation of temperature (albedo and climate sensitivity) and those parameters involved in the uptake of carbon in the surface ocean (ocean surface temperature, salinity, wind stress, and ocean circulation). We selected eight land and ocean parameters, varying each by $\pm 10\%$ relative to the RCP8.5 control, and we compare the percentage change from the reference and the perturbation cases in 2005, 2100, and 2300. The reference, RCP8.5, refers to the tuned set of parameters found in Hector v1.1, resulting in Figures 2-6.

2.3 Data Sources

All RCP input emission data are available at http://tntcat.iiasa.ac.at/RcpDb/. Comparison data are obtained from a suite of CMIP5 Earth System Models (Table 2) (Taylor et al., 2012). The CMIP5 output is available from the Program for Climate Model Diagnostics and Intercomparison (http://pcmci3.llnl.gov/esgct/home.htm). We took the 0-100m (depth) mean for all available CMIP5 data for six output variables, computing the monthly mean for all years in the historical (1850-2005) and RCP 8.5 (2006-2300) experiments. All outputs were regridded to a standard 1-degree grid using bilinear interpolation in CDO version 1.7.1rc1, and then high latitude (-90 to -55 and 55 to 90 degrees), low latitude (-55 to 55), and global area-weighted means computed using R 3.2.4. All CMIP5 comparisons
used in this study are from model runs with prescribed atmospheric CO$_2$ concentrations. We acknowledge that this is not a perfect comparison, as Hector is emissions-forced being compared to the concentration-forced CMIP5 models, but very few of the latter were run under prescribed emissions. We use a combination of root mean square error (RMSE), rates of change ($\Delta$) and bias (degree of systematic over- or underestimation) to characterize Hector’s performance relative to the CMIP5 median.

We also compare Hector to a series of observational ocean data. Surface ocean observations of DIC, pCO$_2$, pH, Ω$_{Ar}$, and Ω$_{Ca}$ are from time-series stations in both the high and latitude oceans; Hawaii Ocean Time Series (HOT), Bermuda Atlantic Time Series (BATS), the European Station for Time Series in the Ocean at the Canary Islands (ESTOC), the Irminger Sea, and the Iceland Sea (Table 3). The time-series data are annually averaged over the upper 100m of the water column. The carbonate parameters not found in Table 3 are computed from temperature, salinity, and the given carbonate parameter pairs using the CO2SYS software (Lewis and Wallace, 1998). The equilibrium constants (K1 and K2 from Mehrbach et al., 1973 refit by Dickson and Millero, 1987) and zero total phosphorus and silica where chosen to best match Hector. Lastly, a longer record (1708 – 1988) of pH and Ω$_{Ar}$ from Flinder’s Reef in the western Coral Sea, calculated from boron isotope measurements, is used in the comparison (Pelejero et al., 2005). We use rates of change ($\Delta$) from 1988-2014, which overlaps the BATS and HOT and BATS time series, to quantify how well Hector simulates the observed changes in the ocean carbonate parameters (Table 6) (Dore et al., 2009; Bates et al., 2014).

3. Results

3.1 Model and Observation Comparisons

Hartin et al. (2015) conducted a thorough analysis of Hector v1.0 demonstrating that it can reproduce the historical trends and future projections of atmospheric [CO$_2$], radiative forcing, and global
temperature change under the four RCPs. In this study we focus on the upper ocean high and low
latitude inorganic carbon chemistry under RCP 8.5.

Hector captures the trend in DIC concentrations for both the high and low latitude surface
ocean with a global RMSE average of 7.0 µmol kg⁻¹ when compared to CMIP5 models over the historical
period (Table 4; Figure 2). We note that there is a systematic bias in both the high and low latitude
surface boxes when compared to CMIP5. First, the carbon pools of the surface boxes are initialized with
carbon values slightly higher than the median CMIP5 values. Second, we note that after 2100 the high
latitude CMIP5 median begins to decline, while Hector rises and stabilizes. Only 3 CMIP5 models ran out
to 2300, with one model driving the decline. Regardless, this offset only results in a <3% global
difference between CMIP5 and Hector.

Hector accurately tracks the pCO₂ in both the high and low latitude surface ocean with similar
rates of change from 1850-2300 (Figure 3). There is a low bias in Hector compared to CMIP5 models
after 2100, due to the low bias in projected atmospheric [CO₂] within Hector over the same time period
(Hartin et al., 2015). We do find Hector to be in closer agreement with the observation record.

Figure 4 shows the high and low latitude surface pH of Hector compared to CMIP5 and
observations from BATS, HOT, ESTOC, Irminger Sea, Iceland Sea, and Flinders Reef. While the high
latitude surface pH is slightly higher than the CMIP5 models, Hector is more similar to high latitude
observations. Since the preindustrial, observations of surface ocean pH decreased by 0.08 units,
corresponding to a 24% increase in [H⁺] concentrations and an 8% decrease in [CO₃²⁻], similar to
numerous studies (Feely et al., 2004; Sabine et al., 2004; Caldeira et al., 2003; Orr et al., 2005) that
estimate an average global decrease in pH of 0.1 or a 30% increase in [H⁺].

The Flinder’s Reef pH record provides a natural baseline to compare future trends in ocean
acidification. While we did not expect the model to match exactly, as this reef site is heavily influenced
by coastal dynamics and internal variability, rates of change from the preindustrial (1750) to 1988 are
the same (0.0002 yr⁻¹) for both Hector and Flinder’s Reef. Over the limited observational record from both the Pacific and Atlantic Oceans, Hector accurately simulates the decline in pH (-0.0017 yr⁻¹) compared to observations (Table 6). Other observations in the North Pacific show surface changes of pH up to 0.06 units between 1991 and 2006 with an average rate of -0.0017 yr⁻¹ (Byrne et al., 2010). Recent work, suggests that the North Atlantic absorbed 50% more anthropogenic CO₂ in the last decade compared to the previous decade, decreasing surface pH by 0.0021 (Woosley et al., 2016). Under RCP 8.5, Hector projects a decrease in low latitude pH of 8.17 in 1850 to 7.77 in 2100 and down to 7.5 by 2300, similar to CMIP5 (Table 5). At approximately 2050, atmospheric [CO₂] is double-twice the preindustrial concentrations, corresponding to a decrease in pH of to 7.96. Shortly after this doubling, pH values are well outside the natural variability found in Flinder’s Reef.

Figure 5, illustrates the high and low latitude surface $\Omega_{\text{Ar}}$. We only highlight $\Omega_{\text{Ar}}$, as $\Omega_{\text{Ca}}$ is similar to that of $\Omega_{\text{Ar}}$. As with pH, Hector is slightly higher than the CMIP5 $\Omega_{\text{Ar}}$ median but closer to the observational record. Hector accurately simulates the change in $\Omega_{\text{Ar}}$ (-0.0090 yr⁻¹) compared to observations (Table 6). Repeated oceanographic surveys in the Pacific Ocean have observed an average 0.34% yr⁻¹ decrease in the saturation state of surface seawater with respect to aragonite and calcite over a 14-year period (1991-2005) (Feely et al., 2012); the average decrease in Hector is between 0.19% yr⁻¹ and 0.25% yr⁻¹. Saturation levels of $\Omega_{\text{Ar}}$ decrease rapidly over the next 100 years in both the high and low latitude. Hector accurately captures the decline in saturations states with low RMSE values for $\Omega_{\text{Ar}}$. Under RCP8.5 Hector projects that low latitude $\Omega_{\text{Ar}}$ will decrease to 2.2 by 2100 and down to 1.4 by 2300. The high latitude oceans will be undersaturated with respect to aragonite by 2100 and will drop down to 0.7 by 2300.

Lastly, Figure 6 highlights pH and $\Omega_{\text{Ar}}$ projections under all four RCPs from 1850 to 2300. Over the last 20 years, both pH and $\Omega_{\text{Ar}}$ have declined sharply and will continue to decline under RCP 4.5, 6.0 and 8.5, outside of their preindustrial and present day values. These RCPs represent a range of possible conditions for the future.
264 future scenarios, with ocean pH varying between 8.15 and 7.46 for the high latitude and Ω_{Ar} varying
265 between 1.94 and 0.60. High latitude Ω_{Ar} saturation levels presently are much lower than the low
266 latitude and reach under-saturation before the end of the century. Even under a best-case scenario, RCP
267 2.6, low latitude pH will drop to 8.077.73 by 2100 and to 8.127.43 by 2300, with Ω_{Ar} saturations states
268 remaining well outside of present day values.

269 3.2 Model Parameter Sensitivity

270 Parametric sensitivities are different between pH and Ω_{Ar}, and between the high and low
271 latitude surface ocean boxes. Parametric sensitivities are different between [H^+] and Ω_{Ar}. We use [H^+] to
272 highlight changes in pH, as pH = -log[H^+]. The reference, RCP8.5, refers to the tuned set of parameters
273 found in Hector v1.1, resulting in Figures 2-6. Global pH is fairly insensitive to the values of the input
274 parameters used, while Ω_{Ar} is slightly more sensitive (Table 7). For example, a 10% change in input
275 parameters results in range from 0.0 - 0.21% in pH and 0.0 - 7.18% in Ω_{Ar}. In comparison a 10%
276 parameter change results in a range from 0.0 - 10.3% in global atmospheric temperature change. In the
277 near term (from 2005-2100) the calculation of pH is sensitive to a combination of parameters, salinity
278 ocean circulation, and beta (terrestrial CO_2 fertilization), and wind stress, while on longer time scales (to
279 2300), [H^+] pH is most sensitive to changes in Q_{10} (terrestrial respiration temperature response) and
280 ocean circulation (Table 7). Global Ω_{Ar} is most sensitive to changes in salinity in both the near and long
281 term. Similar to [H^+] pH, Ω_{Ar} becomes more sensitive to changes in Q_{10} in the long term.

282 4. Discussion

283 The marine carbonate system is projected to undergo significant changes under the RCPs. pCO_2
284 and DIC are increasing rapidly as atmospheric [CO_2] continues to rise under RCP 4.5, 6.0 and 8.5. pH, and
285 Ω_{Ar} are decreasing rapidly outside of observations and are projected to continue to decrease under all
286 scenarios (Figure 6). Only under RCP 2.6 do pH and Ω_{Ar} values begin to increase back towards present
day values. A lowering of $\Omega_{\text{Ar}}$ from approximately 4.0 to 3.0 is predicted to lead to significant reductions in calcification rates in tropical reefs (Kleypas et al., 1999a; Silverman et al., 2009). By the end of the 21st century in agreement with Roy et al., (2015) and Ricke et al., (2013) by the end of the 21st century (2072 under RCP8.5) Hector projects that the low latitude oceans $\Omega_{\text{Ar}}$ will drop below 3.0, well outside of the preindustrial values of $\Omega_{\text{Ar}} > 3.5$, and At the end of the 21st Century, the high latitude oceans $\Omega_{\text{Ar}}$ are close to undersaturation ($\Omega<1$) (Figure 6). These results agree with other studies that investigated changes to the carbonate system (Roy et al., 2015; Ricke et al., 2013). However, the threshold for biogenic carbonate precipitation is species dependent and may be significantly higher than 1.0 when combined with other factors. For example, some coral reef communities need to develop in waters with $\Omega_{\text{Ar}} > 3.3$ (Hoegh-Guldberg et al., 2007; Kleypas et al., 1999b)(Pelejero et al., 2010; Hoegh-Guldberg et al., 2007; Kleypas et al 1999). Accounting for seasonal variations in the $\Omega_{\text{Ar}}$ saturation levels may move the time of under-saturation forward by up to 17 ± 10 years (Sasse et al., 2015). Due to Hector’s time step of 1 year, we may be overestimating the time when ocean acidification reaches a critical threshold. We also note that other factors such as eutrophication, river discharge, and upwelling will likely increase the probability that coastal regions will experience the effects of ocean acidification sooner than the projected open ocean values in Hector (Ekstrom et al., 2015).

Using $[H^+]$ as a proxy for pH, we find that $[H^+]$ pH is sensitive to $Q_{10}$ and ocean circulation, fairly insensitive to most parametric changes, but in both the near and long term, pH is sensitive to parameters that indirectly affect atmospheric $[CO_2]$—Changes in both beta and $Q_{10}$, (the terrestrial CO$_2$ fertilization effect and the respiration temperature response, respectively) are responsible for the uptake and release of carbon within on the land. Uncertainties in the land carbon cycle have been attributed to uncertainties in future CO$_2$ projections within the CMIP5 models (Friedlingstein et al., 2014). Therefore, uncertainties in the land carbon cycle will also have implications for the marine carbonate system. A 10% change in the thermohaline circulation parameter (Tt in Figure 1), representing
a portion of the high latitude to deep ocean circulation, results in ~3% change in $[H^+]$. The dynamics of ocean uptake of $CO_2$ are strongly dependent on this circulation of $CO_2$ laden waters from the surface ocean to depth. CMIP5 models project a weakening in the Atlantic meridional overturning circulation by an average of 36% under RCP8.5 by 2100 (Cheng et al., 2013). Therefore, changes in ocean circulation may have implications on the marine carbonate system, influencing the ocean pH. We also find that the high latitude surface ocean is more sensitive to parameter changes than the low latitude surface ocean. The high latitude box makes up 15% of the global oceans in Hector and therefore, changes of the same size are more easily felt in the high latitude box compared to the low latitude box. This may have direct implications on future marine carbonate projections.

Global $\Omega_{Ar}$ saturation levels are most sensitive to changes in salinity. Within Hector, $\Omega_{Ar}$ is used to directly involved in the calculation $[Ca^{2+}]$ and total boron (Appendix B); is used to determine $\Omega_{Ar}$. Typically the carbonate system is normalized to changes in salinity to understand the chemical changes within the system, instead we show that $\Omega_{Ar}$ may be sensitive not only to future changes in atmospheric $[CO_2]$ but also sensitive to changes in precipitation and evaporation. This may be important, as studies suggest significant changes in precipitation patterns under a changing climate (Held and Soden, 2006; Liu and Allan, 2013).

The dynamics of ocean uptake of $CO_2$ are strongly dependent on the rate of downward transport of $CO_2$ laden waters from the surface ocean to depth. Climate feedbacks on the carbonate system resulting from changes in ocean circulation are neglected in Hector, as the model holds ocean circulation constant in time. CMIP5 models project a weakening in the Atlantic meridional overturning circulation by an average of 36% under RCP8.5 by 2100 (Cheng et al., 2013). We investigate the sensitivity of the carbonate system to a change in ocean circulation by varying the thermohaline circulation parameter ($T_t$ in Figure 1). This parameter represents a portion of the high latitude surface to the deep ocean circulation. A 10% change in ocean circulation ($T_t$) results in a <4% change in air-sea...
fluxes and moderate effects on surface pH and $\Omega_{\text{Ar}}$. If we scale up from a 10\% change in $T$ to a 36\% change, which is projected from the CMIP5 models, it may result in a roughly 14\% change in the air-sea fluxes of carbon to the surface ocean and a 0.3\% and 5.0\% change in pH and $\Omega_{\text{Ar}}$, respectively.

**Conclusions**

We developed a simple, open-source, object oriented carbon cycle climate model, Hector, that reliably reproduces the median of the CMIP5 climate variables (Hartin et al., 2015). The ocean component presented in this study calculates the upper ocean carbonate system ($\text{pCO}_2$, DIC, pH, $\Omega_{\text{Ar}}$, $\Omega_{\text{Ca}}$). Under all four RCPs, pH and $\Omega_{\text{Ar}}$ decrease significantly outside of their preindustrial values matching both observations and CMIP5. In the near future the open ocean and coral reef communities are likely to experience pH and carbonate saturation levels unprecedented in the last 2 million years (Hönisch et al., 2009).

This study is timely because the CMIP5 archive includes a large suite of ESMs that contained dynamic biogeochemistry, allowing us to study future projections of the marine carbon cycle. Rather than running the ESMs, we can use Hector to quickly emulate the global CMIP5 median for projection studies under different emission pathways and sensitivity analyses of the marine carbonate system. **Overall, within this study, we find that numerous parameters influence $[\text{H}^+]$ and $\Omega_{\text{Ar}}$, with both being sensitive to $Q_{10}$, have the most impact on future changes in ocean acidification.** Due to Hector’s simplistic nature and fast execution times, Hector has the potential to be a critical tool to the decision-making, scientific, and integrated assessment communities, allowing for further understanding of future changes to the marine carbonate system.
Appendix A: Model Description – carbon cycle

The carbon component in Hector contains three carbon reservoirs: a single well-mixed atmosphere, a land component and an ocean component. The change in atmospheric carbon is a function of the anthropogenic emissions ($F_A$), land-use change emissions ($F_{LC}$), and atmosphere-ocean ($F_O$) and atmosphere-land ($F_L$) carbon fluxes. The default model time step is 1 year.

$$\frac{dC_{atm}(t)}{dt} = F_A(t) + F_{LC}(t) - F_O(t) - F_L(t)$$  \hspace{1cm} (1)

The terrestrial cycle in Hector contains vegetation, detritus, and soil, all linked to each other and the atmosphere by first-order differential equations. Vegetation net primary production is a function of atmospheric $CO_2$ and temperature. Carbon flows from the vegetation to detritus to soil and loses fractions of carbon to heterotrophic respiration on the way. An ‘earth’ pool debits carbon emitted as anthropogenic emissions, allowing a continual mass-balance check across the entire carbon cycle.

Atmosphere-land fluxes at time $t$ are calculated by:

$$F_L(t) = \sum_{i=1}^{n} NPP_i(t) - RH_i(t)$$  \hspace{1cm} (2)

where $NPP$ is the net primary production and $RH$ is the heterotrophic respiration summed over user-specified n groups (i.e., latitude bands, political units, or biomes) (Hartin et al 2015).

Appendix B: Ocean Carbonate Chemistry

The ocean’s inorganic carbon system is solved via a series of equations modified from Zeebe and Wolf-Gladrow (2001). TA and DIC are used to calculate the other variables of the carbonate system:

$$DIC \times \left(\frac{K_1}{[H^+]} + 2\frac{K_1K_2}{[H^+]^2}\right) = (TA - \frac{K_B}{[H^+]} - \frac{K_W}{[H^+]^2} - [H^+]) \times \left(1 + \frac{K_1}{[H^+]} + \frac{K_1K_2}{[H^+]^2}\right)$$  \hspace{1cm} (1)

This equation results in a higher order polynomial equation for $H^+$, in which the roots (1 positive, 4 negative) are solved for. Once $H^+$ is solved for, pH, $pCO_2$, $HCO_3^-$, and $CO_3^{2-}$ can be determined. We
ignore the nonideality of CO₂ in air and therefore use the partial pressure of CO₂ instead of the fugacity of CO₂. Fugacity is slightly lower by ~0.3% compared to pCO₂ (Riebesell et al., 2009; Sarmiento and Gruber, 2006).

\[ [CO_2] = \frac{DIC}{\left(1 + \frac{K_1}{[H^+]} + \frac{K_1 K_2}{[H^+]^2}\right)} \] (2)

\[ pCO_2 = \frac{[CO_2]}{K_H} \] (3)

\[ [HCO_3^-] = \frac{DIC}{\left(1 + \frac{[H^+]}{K_1} + \frac{K_2}{[H^+]^2}\right)} \] (4)

\[ [CO_3^{2-}] = \frac{DIC}{\left(1 + \frac{[H^+]}{K_2} + \frac{[H^+]^2}{K_1 K_2}\right)} \] (5)

\[ K_1 = \frac{[H^+] [HCO_3^-]}{[CO_2]} \] (6)

\[ K_2 = \frac{[H^+] [CO_3^{2-}]}{[HCO_3^-]} \] (7)

\[ K_B = \frac{[H^+] [B(OH)_4^-]}{[B(OH)_3^-]} \] (8)

\[ K_B \] is the dissociation constant of boric acid from DOE (1994).

\[ B = 416.0 \times \frac{s}{35.0} \] (9)

Total boron is from DOE (1994).

\[ K_W = \frac{[H^+]}{[OH^-]} \] (10)

\[ K_W \] is the dissociation constant of water from Millero (1995).

\[ K_{sp} = [Ca^{2+}] \times [CO_3^{2-}] \] (11)
K$_{sp}$ of aragonite and calcite is calculated from Mucci, (1983).

For those equations with multiple coefficients:

1) K$_H$ and K$_0$ are similar equations calculating Henry’s constant or the solubility of CO$_2$, but they return different units (mol kg$^{-1}$ atm$^{-1}$ and mol L$^{-1}$ atm$^{-1}$) (see Weiss, 1974 for equations and coefficients). K$_H$ is used to solve pCO$_2$ while K$_0$ is used to solve air-sea fluxes of CO$_2$.

2) The Schmidt number is taken from Wanninkhof (1992) for coefficients of CO$_2$ in seawater.

3) [Ca$^{2+}$] (mol kg$^{-1}$) is calculated from Riley and Tongudai (1967).

Acknowledgements

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Author Contributions

C. Hartin designed and carried out the experiments. C. Hartin, B. Bond-Lamberty, and P. Patel developed the model code. A. Mundra processed the data and prepared the figures. C. Hartin prepared the manuscript with contributions from all co-authors.
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<thead>
<tr>
<th>Description</th>
<th>Value</th>
<th>Notes</th>
</tr>
</thead>
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<tr>
<td>Albedo*</td>
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<td>Constant global albedo from 1950 - 2300</td>
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<td>Area of ocean</td>
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<td>Knox and McElroy, (1984)</td>
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<td>Terrestrial CO$_2$ fertilization Wullschleger et al. (1995)</td>
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<td>Sarmiento and Toggweiler, (1984)</td>
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<td>Terrestrial respiration temperature response Davidson and Janssens (2006)</td>
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<td>Thickness of intermediate ocean</td>
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<td>Thickness of deep ocean</td>
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<td>Volume of LL</td>
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<td>Volume of IO</td>
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<td>Salinity HL and LL*</td>
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<td>Initial temperature of LL(^1)</td>
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<td>Thermohaline circulation ((T_t))(^1)</td>
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<td>Wind speed HL and LL(^1)</td>
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<td>Takahashi et al.(^7) [2009]; Liss and Merlivat, (1986)</td>
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</table>

* Parameters contained within the input file.

\(^1\) Parameters varied for the sensitivity analysis.
Table 2: CMIP5 ESM models used in this study containing ocean carbonate parameters. $\Omega_{Ar}, \Omega_{Ca}$ were calculated from the model sea surface temperature, sea surface salinity, and $CO_3$ concentrations.

<table>
<thead>
<tr>
<th>Model</th>
<th>Parameters (RCP 8.5)</th>
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<tbody>
<tr>
<td>BCC-cm1-1</td>
<td>$pCO_2$, temperature</td>
</tr>
<tr>
<td>BNU-ESM</td>
<td>$pCO_2$</td>
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<tr>
<td>CanESM2</td>
<td>DIC, pH, salinity</td>
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<tr>
<td>CESM1-BGC</td>
<td>$CO_3$, DIC, pH, salinity</td>
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<td>CMCC-CESM</td>
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<tr>
<td>CNRM-CM5</td>
<td>$CO_3$, DIC</td>
</tr>
<tr>
<td>GFDL-ESM2G</td>
<td>$pCO_2$, temperature, pH, salinity</td>
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<td>GFDL-ESM2M</td>
<td>$pCO_2$, temperature, $CO_3$, pH, DIC, salinity</td>
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<td>GISS-E2-H-CC</td>
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<td>GISS-E2-R-CC</td>
<td>$pCO_2$, temperature, DIC, salinity</td>
</tr>
<tr>
<td>HadGEM2-CC</td>
<td>$pCO_2$, temperature, $CO_3$, DIC, pH, salinity</td>
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<tr>
<td>HadGEM2-ES</td>
<td>$pCO_2$, temperature, $CO_3$, DIC, pH, salinity</td>
</tr>
<tr>
<td>IPSL-CM5A-LR</td>
<td>Temperature*, $CO_3$, DIC*, pH*, salinity*</td>
</tr>
<tr>
<td>IPSL-CM5A-MR</td>
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<td>IPSL-CM5B-LR</td>
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<td>$pCO_2$, temperature</td>
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<tr>
<td>NorESM1-ME</td>
<td>$pCO_2$, temperature, $CO_3$, DIC, pH, salinity</td>
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* Variable output to 2300.
Table 3: Observational time-series information and carbonate parameters from each location.

<table>
<thead>
<tr>
<th>Time-Series Site</th>
<th>Location</th>
<th>Time-Series Length</th>
<th>Reference</th>
<th>Ocean Carbon Parameters</th>
<th>Data Access</th>
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Table 4: Model validation metrics for the a) high latitude and b) low latitude ocean carbonate variables comparing Hector to CMIP5 from 1850-2005.

<table>
<thead>
<tr>
<th></th>
<th>a) RMSE</th>
<th>b) RMSE</th>
</tr>
</thead>
<tbody>
<tr>
<td>DIC</td>
<td>10.00</td>
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</tr>
<tr>
<td>pCO₂</td>
<td>2.65</td>
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</tr>
<tr>
<td>pH</td>
<td>0.004</td>
<td>0.004</td>
</tr>
<tr>
<td>Ωₐr</td>
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</tr>
<tr>
<td>Ωₖa</td>
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Table 5: Absolute values and rates of change per year (Δ) for the a) high and b) low latitude surface ocean from 1850-2100 and 2101-2300 under RCP 8.5 for DIC (µmol kg⁻¹), pCO₂ (µatm), total-pH (total scale, unitless), Ωₐr (unitless) and Ωₖa (unitless).

<table>
<thead>
<tr>
<th></th>
<th>a) 1850</th>
<th>2100</th>
<th>2300</th>
<th>1850</th>
<th>2100</th>
<th>2300</th>
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<th>2100</th>
<th>2300</th>
<th>1850</th>
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<th>2300</th>
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<tbody>
<tr>
<td>Hector</td>
<td>2107.5</td>
<td>2258.1</td>
<td>2335.5</td>
<td>244.7</td>
<td>816.6</td>
<td>1732.1</td>
<td>8.23</td>
<td>7.76</td>
<td>7.46</td>
<td>2.2</td>
<td>1.0</td>
<td>0.6</td>
</tr>
<tr>
<td>Δ</td>
<td>0.602</td>
<td>0.387</td>
<td>2.29</td>
<td>4.58</td>
<td>-</td>
<td>-</td>
<td>0.0019</td>
<td>0.0015</td>
<td>-</td>
<td>0.0048</td>
<td>-0.002</td>
<td>0.0008</td>
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<tr>
<td>CMIP5</td>
<td>2104.50</td>
<td>2175.79</td>
<td>2243.41</td>
<td>271.62</td>
<td>871.00</td>
<td>1903.82</td>
<td>8.17</td>
<td>7.70</td>
<td>7.38</td>
<td>1.82</td>
<td>0.75</td>
<td>0.44</td>
</tr>
<tr>
<td>Δ</td>
<td>0.285</td>
<td>0.34</td>
<td>2.40</td>
<td>5.16</td>
<td>-</td>
<td>-</td>
<td>0.0019</td>
<td>0.0016</td>
<td>-</td>
<td>0.0048</td>
<td>-0.0003</td>
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<table>
<thead>
<tr>
<th></th>
<th>a) 1850</th>
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<th>1850</th>
<th>2100</th>
<th>2300</th>
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<th>2100</th>
<th>2300</th>
<th>1850</th>
<th>2100</th>
<th>2300</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hector</td>
<td>2073.9</td>
<td>2264.1</td>
<td>2357.6</td>
<td>294.7</td>
<td>879.6</td>
<td>1766.5</td>
<td>8.17</td>
<td>7.77</td>
<td>7.50</td>
<td>4.1</td>
<td>2.2</td>
<td>1.4</td>
</tr>
<tr>
<td>Δ</td>
<td>0.76</td>
<td>0.47</td>
<td>2.34</td>
<td>4.43</td>
<td>-</td>
<td>-</td>
<td>0.0016</td>
<td>0.0014</td>
<td>-</td>
<td>0.0076</td>
<td>0.0040</td>
<td>-0.0116</td>
</tr>
<tr>
<td>CMIP5</td>
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<td>2163.16</td>
<td>2298.89</td>
<td>290.47</td>
<td>930.92</td>
<td>1965.23</td>
<td>8.16</td>
<td>7.73</td>
<td>7.45</td>
<td>3.75</td>
<td>2.00</td>
<td>1.36</td>
</tr>
<tr>
<td>Δ</td>
<td>0.66</td>
<td>0.68</td>
<td>2.56</td>
<td>5.17</td>
<td>-</td>
<td>-</td>
<td>0.0011</td>
<td>0.0014</td>
<td>-</td>
<td>0.0070</td>
<td>0.0032</td>
<td>-0.0049</td>
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</table>
Table 6: Trends and standard error for the carbonate system taken from Bates et al., (2014) and values calculated from Hector and CMIP5. *Global carbonate system parameters for Hector and CMIP5 from 1988-2014.*

<table>
<thead>
<tr>
<th></th>
<th>Length of record</th>
<th>DIC (µmol kg⁻¹ yr⁻¹)</th>
<th>pCO₂ (µatm yr⁻¹)</th>
<th>pH (yr⁻¹)</th>
<th>ΩAr (yr⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BATS</td>
<td>1983-2014</td>
<td>1.37 ± 0.07</td>
<td>1.69 ± 0.11</td>
<td>–0.0017 ± 0.0001</td>
<td>–0.0095 ± 0.0007</td>
</tr>
<tr>
<td>HOT</td>
<td>1988-2014</td>
<td>1.78 ± 0.12</td>
<td>1.72 ± 0.09</td>
<td>–0.0016 ± 0.0001</td>
<td>–0.0084 ± 0.0011</td>
</tr>
<tr>
<td>ESTOC</td>
<td>1995-2014</td>
<td>1.09 ± 0.10</td>
<td>1.92 ± 0.24</td>
<td>–0.0018 ± 0.0002</td>
<td>–0.0115 ± 0.0023</td>
</tr>
<tr>
<td>Iceland Sea</td>
<td>1983-2014</td>
<td>1.22 ± 0.27</td>
<td>1.29 ± 0.36</td>
<td>–0.0014 ± 0.0005</td>
<td>–0.0018 ± 0.0027</td>
</tr>
<tr>
<td>Irminger Sea</td>
<td>1983-2014</td>
<td>1.62 ± 0.35</td>
<td>2.37 ± 0.49</td>
<td>–0.0026 ± 0.0006</td>
<td>–0.0080 ± 0.0040</td>
</tr>
<tr>
<td>Hector</td>
<td>1988-2014</td>
<td>0.90</td>
<td>1.82</td>
<td>–0.0017</td>
<td>–0.0089</td>
</tr>
<tr>
<td>CMIP5</td>
<td>1988-2014</td>
<td>0.68</td>
<td>1.77</td>
<td>–0.0018</td>
<td>–0.0074</td>
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</tbody>
</table>
Table 7: Percentage change from reference (RCP8.5) for two Hector outputs a) global $[\text{H}^+]\text{pH}$ and b) global $Q_{Ar}$, for a ±10% change in eight model parameters. Results are shown for three years, 2005, 2100 and 2300.

<table>
<thead>
<tr>
<th>a) Year</th>
<th>Parameter</th>
<th>+10% change</th>
<th>-10% change</th>
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<tbody>
<tr>
<td>2005</td>
<td>Albedo</td>
<td>0.13</td>
<td>0.00</td>
</tr>
<tr>
<td>2005</td>
<td>Beta (terrestrial CO$_2$ fertilization)</td>
<td>0.63</td>
<td>-0.50</td>
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<tr>
<td>2005</td>
<td>Ocean Circulation (T$_{1}$)</td>
<td>0.76</td>
<td>-0.76</td>
</tr>
<tr>
<td>2005</td>
<td>$Q_{40}$ (terrestrial respiration temperature response)</td>
<td>-0.13</td>
<td>0.13</td>
</tr>
<tr>
<td>2005</td>
<td>Salinity</td>
<td>-0.88</td>
<td>1.51</td>
</tr>
<tr>
<td>2005</td>
<td>Climate Sensitivity</td>
<td>-0.13</td>
<td>0.13</td>
</tr>
<tr>
<td>2005</td>
<td>Surface ocean temperature</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>2005</td>
<td>Wind Stress</td>
<td>-0.38</td>
<td>0.63</td>
</tr>
<tr>
<td>2010</td>
<td>Albedo</td>
<td>0.59</td>
<td>0.00</td>
</tr>
<tr>
<td>2010</td>
<td>Beta (terrestrial CO$_2$ fertilization)</td>
<td>1.78</td>
<td>-1.78</td>
</tr>
<tr>
<td>2010</td>
<td>Ocean Circulation (T$_{1}$)</td>
<td>2.37</td>
<td>-1.78</td>
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<tr>
<td>2010</td>
<td>$Q_{40}$ (terrestrial respiration temperature response)</td>
<td>-0.59</td>
<td>1.18</td>
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<tr>
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<td>Salinity</td>
<td>1.88</td>
<td>-1.25</td>
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<tr>
<td>2010</td>
<td>Climate Sensitivity</td>
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<td>2.50</td>
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<tr>
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<td>Surface ocean temperature</td>
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<td>0.59</td>
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<td>2010</td>
<td>Wind Stress</td>
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<td>1.78</td>
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<td>2030</td>
<td>Albedo</td>
<td>0.00</td>
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<td>2030</td>
<td>Beta (terrestrial CO$_2$ fertilization)</td>
<td>1.56</td>
<td>-1.88</td>
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<tr>
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<td>Ocean Circulation (T$_{1}$)</td>
<td>2.81</td>
<td>-3.44</td>
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<td>$Q_{40}$ (terrestrial respiration temperature response)</td>
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<td>3.44</td>
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<tr>
<td>2030</td>
<td>Salinity</td>
<td>1.88</td>
<td>-1.25</td>
</tr>
<tr>
<td>2030</td>
<td>Climate Sensitivity</td>
<td>-2.5</td>
<td>2.50</td>
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<tr>
<td>2030</td>
<td>Surface ocean temperature</td>
<td>0.62</td>
<td>0.31</td>
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<tr>
<td>2030</td>
<td>Wind Stress</td>
<td>-1.25</td>
<td>1.25</td>
</tr>
<tr>
<td>Year</td>
<td>Parameter</td>
<td>+10% change</td>
<td>-10% change</td>
</tr>
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<td>-----------</td>
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</tr>
<tr>
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<td>Albedo</td>
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<td>-0.00</td>
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<td>2300</td>
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<td>0.00</td>
</tr>
<tr>
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<td>Beta (terrestrial CO$_2$ fertilization)</td>
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<td>-1.34</td>
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<td>-1.35</td>
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<td>2300</td>
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<tr>
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</tr>
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<td>3.00</td>
</tr>
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<td>Salinity</td>
<td>3.80</td>
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<td>2100</td>
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<td>-5.89</td>
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<td>-7.18</td>
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<tr>
<td>2005</td>
<td>Climate Sensitivity</td>
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<td>-0.07</td>
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<td>2100</td>
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<td>0.55</td>
<td>-0.56</td>
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<tr>
<td>2300</td>
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<td>0.43</td>
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<td>2005</td>
<td>Surface ocean temperature</td>
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<td>-1.99</td>
</tr>
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<td></td>
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<tr>
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<td>-2.27</td>
</tr>
<tr>
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<td>Wind Stress</td>
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<td>0.25</td>
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<td>2100</td>
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<td>0.88</td>
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<tr>
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<td></td>
<td>-1.13</td>
<td>0.88</td>
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</table>
Figure 1: Representation of the carbon cycle in Hector. The atmosphere consists of one well-mixed box, connected to the surface ocean via air-sea fluxes of carbon. The terrestrial component consists of user-defined biomes or regions for vegetation, detritus, and soil. The earth pool is continually debited to act as a mass balance check on the carbon cycle (Hartin et al., 2015). The ocean consists of four boxes, with advection (represented by solid straight arrows) and water mass exchange (represented by dashed curved arrows) simulating thermohaline circulation. The marine carbonate system is solved for in the high and low latitude surface boxes. At steady state, there is a flux of carbon from the atmosphere to the high latitude surface box, while the low-latitude surface ocean releases carbon to the atmosphere.
Figure 2: Dissolved inorganic carbon (DIC) for high (top) and low latitude (bottom) surface ocean under RCP 8.5; Hector (blue), CMIP5 median, standard deviation, and model range (red, n = 15 (1850-2100, with n=4 starting after 1860) and n= 3 (2101-2300)); and observations from BATS (teal), ESTOC (pink), HOT (purple), Iceland (yellow) and Irminger Sea (navy). Note a doubling of CO$_2$ from preindustrial values occurs around 2050.
Figure 3: $pCO_2$ for high (top) and low latitude (bottom) surface ocean under RCP 8.5; Hector (blue), CMIP5 median, standard deviation, and model range (red, $n = 15$ (1850-2100) and $n= 2$ (2101-2300)); and observations from BATS (teal), HOT (purple), ESTOC (pink), Iceland (yellow) and Irminger Sea (navy).
Figure 4: pH for high (top) and low latitude (bottom) surface ocean under RCP 8.5; Hector (blue), CMIP5 median, standard deviation, and model range (red, $n = 13$ (1850-2100) and $n = 2$ (2101-2300)); and observations from BATS (teal), ESTOC (pink), HOT (purple) Flinder’s Reef (green), Iceland (yellow) and Irminger Sea (navy).
**Figure 5:** Aragonite saturation ($\Omega_{Ar}$) for high (top) and low latitude (bottom) surface ocean under RCP 8.5; Hector (blue), CMIP5 median, standard deviation, and model range (red, $n = 10$ (1850-2100) and $n=2$ (2101-2300)); and observations from BATS (teal), HOT (purple) and Flinder’s Reef (green).
Figure 6: High and low latitude a) pH and b) aragonite saturation ($\Omega_{Ar}$) time series for Hector from 1850-2300 for RCP 2.6 (red), RCP 4.5 (green), RCP 6.0 (teal) and RCP 8.5 (purple). Note that even under a strongly mitigated scenario (RCP 2.6), both $\Omega_{Ar}$ and pH at 2300 are still lower than preindustrial values.
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