Replies to Reviewer #2

We thank the reviewer for their critical assessment, very constructive comments and suggestions, as well as their positive overall assessment of our manuscript. We followed or otherwise implemented most reviewer comments. Below we explain how we will address every suggestion during the revision of the manuscript. The reviewer's comments are in blue and our replies in black.

Suggestions General

1. pH and pCO_2 I see in the literature it is mixed for pCO_2 vs pCO_2 (I don't quite understand why it is sometimes given italics?). I don't think I have previously seen italics for pH (plus the two p's mean two different things). I generally argue for partial pressure of CO_2 to be rendered as PCO_2 to match with P as the symbol for pressure (though I acknowledge the double subscript is typographically annoying) and atmospheric CO_2 concentration (in ppm) as simply "atmospheric CO_2". I leave it up to you which form you use.

Thank you for pointing this out. We will go for the non-italicised small p for pH and used 'atmospheric CO_2' throughout the manuscript.

2. Equilibrium Constants I see that you have caveated that 'equilibrium constants' (and Ks) refers to stoichiometric/ apparent equilibrium constants on line 176 - but there are several mentions and equations before this (including in the abstract). The terminology within the field is frustrating. My understanding is that 'equilibrium constants' (the true ones, that are calculated using activity) really are constant and are generally given the symbol K. Incorporation of the activity coefficients into those termsgives us 'apparent equilibrium constants' (often given the symbol K*) which are poorly named because despite having 'constants' in the name they're not constant at all. The general confusion is promulgated by 'apparent equilibrium constants' being shortened to have the same name equilibrium constants. I think it would be clearer to refer to them as apparent equilibrium constants with the symbol K* throughout, and use K* or K*s in the text if you want to avoid having a lengthy phrase repeated many times.

We will change all Ks to K*s as suggested.

3. Figures are low resolution throughout Particularly noticeable for Figures 2, 3, 4, and 6 (zooming to look at an individual subplot).

We apologise for the low resolution. We will attach the high-resolution pdf versions of all figures to the revised manuscript

Targeted suggestions

4. Lines 74-80: There's a list of three things 1) ionic strength, 2) interactions with carbon and boron, 3) complexation. Could you clarify if 2) and 3) are different things (as opposed to 3) being a specific type of 2))? That clarification needn't be in the manuscript if the distinction is clear to someone more familiar than me.

This listing summarizes an escalation: the direct "ideal" ionic strength effect from extended Debye-Hückel theory is very weak, the effect of "non-ideal" ion-ion interactions with Mg and Ca on free-ion activity coeficients are also weak, and finally it is primarily the complexation of the majority of total carbonate ion by Mg and Ca that affects the fraction of total carbonate ion that is free. For Eocene seawater, this latter effect on pK2 and pKsp is an order of magnitude stronger than the sum of the former effects.
We will clarify that 3) is an explanation of 2).

5. Line 85-86: The final line of this paragraph felt a little out of place (it gave me the impression the next paragraph was going to introduce PITZER models). I think it could go in line 82 just after you cite Hain et al 2015?

We will follow the suggestion and move this sentence to line 82.

6. Lines 224 - 226: You mention that the K*s for sulfate and fluoride are on the seawater scale - is that right? And if so, how is that done?* Ks for sulfate and fluoride are typically left on the free scale because they're involved in the free -> total -> seawater scale calculation. To put the K* of sulfate from the free onto the total/seawater scale, you need to know how much HSO_4^- and SO_4^{2-} are in the solution, which is typically achieved by prescribing the total sulfur concentration, then using K* for sulfate to calculate the speciation. It could work if speciated products are prognostic variables from the model? Otherwise, I think you end up in a loop where you need the K* to calculate speciation to calculate the K* to calculate the speciation.

The carbonate solver in the model performs the first loop referred to by the reviewer: It first estimates the sulphate and flouride speciation using K*s on the free scale and assumed total dissolved sulphate and flouride. Furthermore, for the pH scale conversion, we assume the modern empirical relationship between salinity and total dissolved sulphate and flouride. These speciations are used to determine all the required conversion factors to the seawater scale. This includes the K*s of sulphate and fluoride, but these adjusted, seawater-scale values are only used in the calculation of carbonate alkalinity. We will add a note to the manuscript to clarify this.

We will aim to improve this in the future and avoid the need to convert scales (this was done in the original Ridgwell [2001] scheme by necessity and in light of what

empirical constants were available at the time).

7. Section 2.4: Describes setting up the experiments, but starts with quite a few lines of highly technical details regarding seasonality and PIC:POC ratios before we get to what the experiments actually are, and what you've tested. As a reader, I feel this may be clearer if you flipped the section round to say something like (paraphrasing): "There are two main facets of the cGENIE carbonate system we wish to test:
   1. Is the lookup table approach accurate enough?
   2. How much does improving the K* calculation affect carbon cycle dynamics? In order to answer these questions, we designed a suite of simulations using different sets of K*s, which we split into three main categories: A) Mehrbach constants without any adjustment for changing seawater major ion chemistry B) Mehrbach constants with major ion chemistry correction of Yaakov and Goldhaber and Tyrrell and Zeebe (the current cGENIE model default) C) MyAMI derived constants Each category was run for both preindustrial and early Eocene seawater chemistry (see the full list in Table 1). To estimate the accuracy of the lookup table approach, we carefully chose a non-seasonal configuration (which minimizes the influence of non-linearities driven by seasonal sea ice extent), and also imposed a fixed pattern of CaCO :POC export by using the annual mean (which removes the carbonate compensation feedback). This ensures we isolated the impact of changingthe method of K* calculation. To understand how changing K* calculation routine affects the dynamic carbon cycle…" In other words putting the headline information at the top of the section, and the (necessary and useful) technical details of precisely which simulations and which configurations slightly later. You've already given this information in other sections, it's just a reiteration as a reminder and I think it will also make the sections dovetail nicely together (for instance it will then match exactly with the first sentence of your results section).

Thank you for this suggestion. We will restructure our experiment design description following the suggested outline. We will first explain the experiments that we performed to assess the accuracy of our look-up table approach and then explain our experiments that explore the implications of the model update. We will provide all technical information, most of which applies to all experiments, at the end of the Method section.

8. Table 1 - Is there a reason that the simulations are in this order? As in, the A, B and C. I would suggest sorting them in the order of: A) No correction, B) Current correction C) Lookup table correction (C2) Lookup table correction (temperature limited)) if it isn't a massive hassle.

There is no need to keep this order. We will change the order as suggested and rename the experiments accordingly

9. Section starting line 386 - suggest adding some subtitles or equivalent to break up this paragraph.

We will break up the paragraph with line breaks and shorten it by reducing repetition.

10. Section 3.2 - the paragraph beginning on line 488 oscillates between observation and interpretation. I would suggest separating the results (what's happening with K*s, pH, etc. in the Arctic/Atlantic/Pacific) from the why (high latitude strongly influenced by temperature limitations, Atlantic/Pacific primarily driven by…). Or separating this into subsections each dealing with a specific area.

We broke up this section with line breaks too and revised it to reduce the jumping between geographic areas

11. Section 3.3 starts with an excellent and very clear description of the impact of changing major ion chemistry, which I think would be better earlier in the manuscript.

We moved this section to the introduction.

12. In Figures 3 and 6 - is it an option for ΔK* maps to be shown using a percentage of the value in the upper plot? It looks to me like they're just about the right order of magnitude that that might work well and would simplify interpretation for non experts.

We followed the suggestion and replaced the difference plots in these figures with plots of percentage changes

13. Line 512+522: "Total DIC in the ocean is 24 PgC higher". Would it be possible to give this as a percentage (or give the total, or give in units of concentration for average seawater DIC change)? I think that would make it easier to interpret in a glance.

We followed the suggestion and added percentages.

14. Paragraph beginning line 611: There are a lot of interesting results here, but they're quite densely packed and from a lot of different simulations. Would a small table work for easy cross comparison of the simulations? Or something to that effect so it is straightforward to parse and compare ΔDIC in EE-C-Open vs EE-B-Open etc.

A full comparison of all these results is already shown in Fig. 7, but we have added the values to the plot to convey the same information as would be in the table. We also added more references to subpanels of the figure to the text to make the connection clearer

Typographical amendments (I've used green for suggested additions and red strikethrough for suggested deletions).

15. Figure 1: Add Mg/Ca to the figure legend. Make the y axes tick labels on the right grey to match the Mg/Ca label. In the figure caption, put the charges on the ions into superscript.

Thank you for spotting this, we will fix it.

16. Line 60: "compared to 52 mmol kg −1 today, (Timofeeff et al. [2006], Brennan et al. [2013], Hain et al. [2015]))." (added brackets around passive citation).

We will correct this.

17. Line 60-63: "The first order consequence of higher Cretaceous [Ca2+] (as we will illustrate later) is that to produce a relatively similar ocean surface carbonate saturation state (or global weathering rate) to the present day, the ocean carbonate ion (CO32-) concentration would have had to have been lower as carbonate saturation involves the product of both [Ca2+] and [CO32-]" (to keep the sentence in the conditional tense) Or alternatively if you could put them both into the non conditional: "The first order consequence of higher Cretaceous [Ca2+] (as we will illustrate later) is that the ocean carbonate ion (CO32-) concentration would have been lower than the present day, assuming a similar ocean surface carbonate saturation state (or global weathering rate)".

We will simplify the sentence as suggested.

18. Line 66: "(e.g., Henehan et al. [2019])" (has a space in the bracket just before 2019).

We will correct this.

19. Line 143: “cbsyst (Branson et al. [2023]), and KGENgen (Whiteford et al. [2025])” (extra n in Branson and decapitalised GEN).

We will correct this.

20. Line 206: “contribution from $SO4^{2-}$” (I think that’s correct it should be sulfate right?)

We intended to write about sulfide (S2-) here, which can contribute to alkalinity at low pH or reducing conditions (Dickson et al. 1981).

21. Line 240: “in the case of methanotrophy (Reinhard et al. [2020]).” (closed the bracket at the end of the paragraph).

We will correct this.

22. Line 242: “Dissolved Ca2+ and total SO_4^{2-} (HSO_4^- + SO_4^{2-}) are typically” (removed the O_4^{2-} to make it total sulfur is sulfate plus bisulfate).

We will correct this.

23. Line 260: “concentrations is < 1 µatm (Ridgwell 260 [2001]).” (closed the bracket after citation).

We will correct this.

24. Lines 264 and 269: WΩ.

We will correct this.

25. Lines 277 and 279: DΔ.

We will correct this.

26. Line 360: “(270 pmµatm/atm pCO_2)” (I think the original pm is probably a typo meant to be ppm? If it’s partial pressure it should technically be in µatm/atm, though the difference between that and ppm is irrelevant for this work).

We will correct this.

27. Line 367: “Tyrrell and Zeebe (2004)” (extra r in Tyrrell).

We will correct this.

28. Line 536: “alkalinity from the model) theimpacts on carbonate” -> “alkalinity from the model) the impacts on carbonate” (added a space in the impacts).

We will correct this.

29. Figures SI.2 and SI.3 are missing K in the label.

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References

Dickson, A.G., 1981. An exact definition of total alkalinity and a procedure for the estimation of alkalinity and total inorganic carbon from titration data. *Deep Sea Research Part A. Oceanographic Research Papers*, *28*(6), pp.609-623.

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1. Line 66: "(e.g., Henehan et al. [2019])" (has a space in the bracket just before 2019).

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2. Line 143: "cbsyst (Branson et al. [2023]), and KGENgen (Whiteford et al. [2025])" (extra n in Branson and decapitalised GEN).

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