Dear editor,

I will be submitting an edited version of the manuscript, "Calcium carbonate dissolution rate changes under future climate scenarios," to *Biogeosciences*. I have changed much of the text and presentation of the figures according to the reviewer's suggestions.

1. I now include a comparison of the calcium carbonate dissolution rate estimates from the present-day period of the CMIP6/GeoMIP output and those from the GLODAPv2 and other observationally-derived data. These estimates compare quite well, suggesting that CMIP6/GeoMIP output has sufficient fidelity to be used to make projections about future calcium carbonate dissolution rates.

2. I also demonstrate that, while there is a shift in potential alkalinity towards lower values in the future climate period relative to the present-day period and there is a change in slope between alkalinity and salinity from the present-day to future climate periods, the slope of the relationship between potential alkalinity and mean age does not significantly change from the present-day period to the future climate period. This suggests the Alk*-mean age slope-based method I use to estimate future calcium carbonate dissolution rates should be about as valid as Sulpis et al. (2021)'s estimates.

3. Lastly, because I increased the uncertainty in the mean ages in the GeoMIP calculations by propagating the error in the mean age associated with not having ages less than 200 years (only having Carbon-14-based ages) and I realized that the units were not consistent in the tracers that should be in $\mu molkg^{-1}$ (instead of $molm^{-3}$), the figures with GeoMIP results have changed as well. These issues were not present with the CMIP6 analyses, but because alkalinity became a significant factor in es-

timating calcium carbonate dissolution rates, I devised a method to predict what the future alkalinity will be based on present-day salinity and alkalinity and future salinity. I compared a Generalized Additive Model-based prediction with a Random Forest-based prediction and found that the Random Forest estimated the future alkalinity within a $10^{-3}\%$ error on average and within a 2.94 $\mu molkg^{-1}$ root-mean-square error. Then I re-estimated the calcium carbonate dissolution rates using the CMIP6 output and these future alkalinity estimates.

I address each of the reviewer's points below.

1 Reviewer #1 (Comments to Author (shown to authors):

- This manuscript investigates how calcium carbonate ($CaCO_3$) dissolution rates in the ocean may change under future climate scenarios, using CMIP6 (SSP5-8.5) and GeoMIP model outputs combined with the diagnostic approach of Sulpis et al. (2021). The study identifies salinity and mean water age as the dominant factors controlling future dissolution rate changes and compares the effectiveness of various geoengineering interventions. While the topic is timely and relevant, given the importance of calcium carbonate dissolution for ocean carbon and alkalinity cycles, the manuscript suffers from several significant weaknesses that limit its scientific contribution.

- **Thank you for taking the time to provide such feedback. I have substantially modified the manuscript in the ways described above and respond**

to each of your concerns below.

- My primary concerns: 1. The manuscript attempts to address too many loosely connected topics (dissolution rates, multiple geoengineering scenarios, sea level, suspended sediments in river deltas) without adequately integrating them into a unified narrative. Section 4 on "Direct vs indirect climate consequences" feels particularly disconnected from the main analysis.

- **Olivier Sulpis and I have performed back-of-the-envelope estimates of how much sea-level has changed since pre-industrial times due to calcium carbonate dissolution (about 84 mm), but Olivier also suggested removing any sea-level analysis when we spoke. Because my current role focuses on sea-level, I heavily edited what I wrote. The connection between the future calcium carbonate dissolution rates is that 40% of sediments are made of calcium carbonate, sediments in sufficiently high concentrations will alter sea-level by Archimedes' principle, and increased dissolution of calcium carbonate-based sediments in the future will therefore contract sea-level due to the reduction in density of the sediment when the calcium carbonate dissolves. This may eventually be a significant factor to account for in regional sea-level budgets, which is the reason why I included this analysis in the manuscript. I focused on river deltas because these are the regions where there are sufficiently high concentrations of sediments to impact sea-level. But I have now removed this discussion from the manuscript. As for the geoengineering scenarios, one of the purposes of my study is to examine the possible future ranges of calcium**

carbonate dissolution rates. The GeoMIP simulations' output serves as a lower bound and the CMIP6 scenario's output serves as an upper bound. I did not include other scenarios because I did not want to broach the question of which scenario is most realistic. I was only interested in the range and understanding the estimates' sensitivities to particular factors.

- 2. The perturbation-based approach, while computationally efficient, is crude and lacks validation. The study does not verify that applying climate model perturbations to present-day observations produces physically meaningful results.

- **Sulpis et al. (2021) performed the validation relative to sediment trap data for the calcium carbonate dissolution rate estimates from the Alk*-mean age slope method they used and I use here. The CMIP6 biogeochemical output has been compared with independent observational data products, at least in the Southern Ocean, by Cheng, M., N. Maher, M. J. Ellwood, 2025: Evaluating the performance of CMIP6 models in simulating Southern Ocean biogeochemistry. Biogeosciences, 22, 7269–7291, 2025 https://doi.org/10.5194/bg-22-7269-2025. In addition, I now compare the calcium carbonate dissolution rate estimates using the same GLODAPv2 and other observationally-derived data that Sulpis et al. (2021) did and those using the near-present-day years of the CMIP6/GeoMIP simulation output. The correlation between these two dissolution rate estimates globally is about 0.93. A figure is shown later in this response. While potential alkalinity using these two data sets has a correlation of about 0.8, it is the slope of the potential alkalinity**

versus mean age relationship that matters, which is why the correlation between the calcium carbonate dissolution rate estimates can be higher than the correlation between the potential alkalinity estimates. If there are other comparisons you would like to see using available data, I am open to suggestions.

- 3. Many key claims about implications (e.g., for sea level, carbon budgets) are left unquantified and deferred to "future studies," which undermines the practical value of this work.

- **The sea-level discussion has now been removed from the manuscript, which is mostly (if not entirely) what you're referring to here. As you seemed to imply above, the sea-level discussion belongs in its own separate manuscript, where I should address the issues that I deferred to future studies.**

- 4. The finding that salinity dominates dissolution rate changes appears to be largely a mathematical artifact of the Alk* formula (Eq. 1), where salinity has a coefficient that is much larger than the other terms.

- **Yes, in the previously submitted draft of the manuscript, I explained why the calcium carbonate dissolution rates are most sensitive to salinity and mean ages mathematically. The underlying issue here is whether the Alk*-mean age slope-based method is valid in a future climate, and not just in the past/present climate in which it was empirically derived. Because this cannot directly be tested (using future observational data that**

doesn't exist yet), I made an attempt to be clear about my assumption that this method is valid in a future climate. To assess this assumption, I have looked at the phase space of alkalinity, nitrate, and salinity (the variables that explicitly come into the Alk* formula). The phase spaces for the present-day and future climate periods are now shown (see later in this response). The main issue with using this method of calculating the calcium carbonate dissolution rates is the extent to which cross-isopycnal transports and mixing become more or less important in a future climate. According to the studies cited in Figure 2.2 of Melet, A. V., R. Hallberg, D. P. Marshall, 2022: Chapter 2 - The role of ocean mixing in the climate system, Editor(s): Michael Meredith, Alberto Naveira Garabato, Ocean Mixing, Elsevier, 5-34, https://doi.org/10.1016/B978-0-12-821512-8.00009-8, there can be regional differences in ocean mixing of up to 40%, but only 5% globally. Together with the strong relationship between our estimates using observationally-derived data and using present-day climate model output, we suggest that the estimates from Sulpis et al. (2021) are approximately as valid over basin scales as our calculations in a future climate context.

- Specific Comments 1. Scope and focus The manuscript covers calcium carbonate dissolution, CMIP6 projections, GeoMIP geoengineering scenarios, sea level implications, and suspended sediment concentrations in river deltas. These topics are not well integrated. Specifically: What is the logical connection between open-ocean $CaCO_3$ dissolution rates and suspended sediment concentrations in river deltas?

The abstract mentions "sea level" implications, but the quantitative link between dissolution rate changes and sea level is never established. I recommend the author either (a) focus the manuscript solely on dissolution rate sensitivity analysis, or (b) provide a much clearer and quantitative framework connecting all topics.

- **The connection between open-ocean calcium carbonate dissolution rates and suspended sediment concentrations is likely negligible. It's the connection between near-coastal calcium carbonate dissolution rates and suspended sediment concentrations that may be important to take into account for regional sea-level budgets applications, but because only 10% of dissolution occurs on the continental shelves (not even the near-coastal environment), the connection is tenuous. I will remove the sea-level component of the manuscript and focus solely on future dissolution rates.**

- 2. Perturbation approach The approach of adding climate model perturbations to observational data (line 37-40) is a significant simplification. The authors acknowledge this but do not adequately address: Why is it valid to assume that the Alk*-age relationship (and its slope) remains unchanged when perturbations are applied? Cross-isopycnal transports and mixing are not accounted for (line 283-284). How large might this error be? The empirical formula for inferring ages from Carbon-14 (Eq. 2) is described as "crude" (line 289) and can produce unreliable results for young waters. This is a major limitation for the GeoMIP analysis. The author should provide at least a qualitative assessment of the errors introduced by these assumptions.

- **The Alk*-mean age relationship is valid within the range of observed**

combinations of environmental properties (temperature, salinity, nitrate, phosphate,...). We now include a phase space diagram that shows that the ranges of the variables explicitly involved in the Alk* equation (alkalinity, nitrate, and salinity) do not significantly change from the present-day simulation output and the future climate output. While temperature does significantly change and the relationship between alkalinity and salinity has a slightly different slope, temperature and alkalinity do not impact Alk* by nearly as much as salinity, which does not change in its slope with mean age in our calculations from the present-day to future climate output. Only the intercept is altered going from the present-day period to the future period because there is a shift to smaller Alk*.

- **The error analysis is a good suggestion and I have attempted to perform this error analysis. For instance, by excluding the mean age perturbations of water less than 200 years old (but still accounting for the Carbon-14-based mean age perturbations) in the GeoMIP output, there is on average a 15.9% difference in the mean age. We now account for this in the calculations by using $\sqrt{0.2^2 + 0.159^2}$ as the total uncertainty in mean age (i.e., assuming these two sources of uncertainty are independent). The cross-isopycnal transport and mixing was already included in the uncertainties in the calculations performed by Sulpis et al. (2021) and myself as a 6 $\mu molkg^{-1}yr^{-1}$ uncertainty in the calcium carbonate dissolution rates. However, the basin-wide comparisons shown in Figure 1 of Wu et al. (2025) between their estimates that account for cross-**

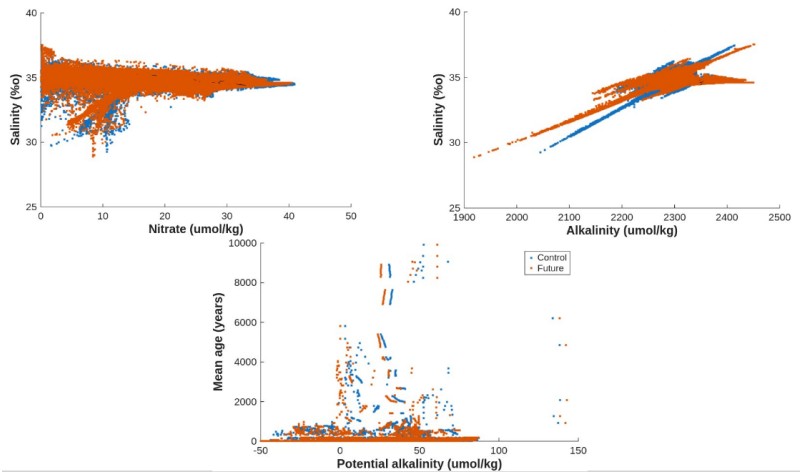

Figure 1: Shown are phase space diagrams between nitrate and salinity (top-left panel), alkalinity and salinity (top-right panel), and Alk* and mean age (bottom panel) over the present-day simulation period of the present-day (blue dots) and the future climate period (orange dots). The slope of the relationship between alkalinity and salinity is smaller under future climate conditions and there is a shift to smaller Alk* values.

isopycal transport and mixing and the estimates of Sulpis et al. (2021) suggest that there is good agreement between the two methods aside from the shallow (above the aragonite saturation depths $< 500 - 1000$ m) subtropical (plus one subpolar) ocean basins, but the uncertainties in the Sulpis et al. (2021) estimates were excluded from their comparison. Because ocean mixing is expected to increase by a small amount globally (5%), we suggest our future climate estimates are valid everywhere Figure 1 of Wu et al. (2025) shows good agreement and place the caveat on our estimates in shallow subtropical (plus the subpolar North Atlantic) ocean basins that our estimates are likely too large when cross-isopycnal transport and mixing are accounted for.

- 3. Model validation The authors state (line 59-61): "We do not perform an assessment of the CMIP6 output with the GLODAPv2 data." While the temporal mismatch is a valid concern, the complete absence of any validation is problematic. Can the authors at least demonstrate that the CMIP6 control simulations produce reasonable dissolution rates compared to Sulpis et al. (2021) estimates, even if the time periods differ?

- **This is also a good suggestion. I have estimated the calcium carbonate dissolution rates using the CMIP6/GeoMIP control simulation output to compare with those estimated with the GLODAPv2 data. They compare fairly well with each other with a correlation of** 0.93**.**

- 4. Salinity dominance could be mathematical artifact? The main finding that salinity dominates future dissolution rate changes (e.g., line 266) needs to be critically examined. Looking at Equation 1: $Alk* = TA + 1.26[NO]66.4S$ The coefficient for salinity (66.4) is $50\times$ larger than for nitrate (1.26) and effectively determines Alk* changes. The authors even acknowledge this (line 114-116): "The impact of salinity on Alk* is apparent through Eq (1)... this gets magnified after scaling those factors by their coefficients." Is the salinity dominance a genuine physical insight or simply a consequence of the formula's structure? I request the author discuss this issue explicitly and clarify what new physical understanding this result provides beyond what is already embedded in the Sulpis et al. (2021) methodology.

- **Yes, I tried to make this clear that the salinity dominance is a direct consequence of the formulation that Carter et al. (2014) developed. The dominance of mean age comes from the slope of the mean age-Alk* re-**

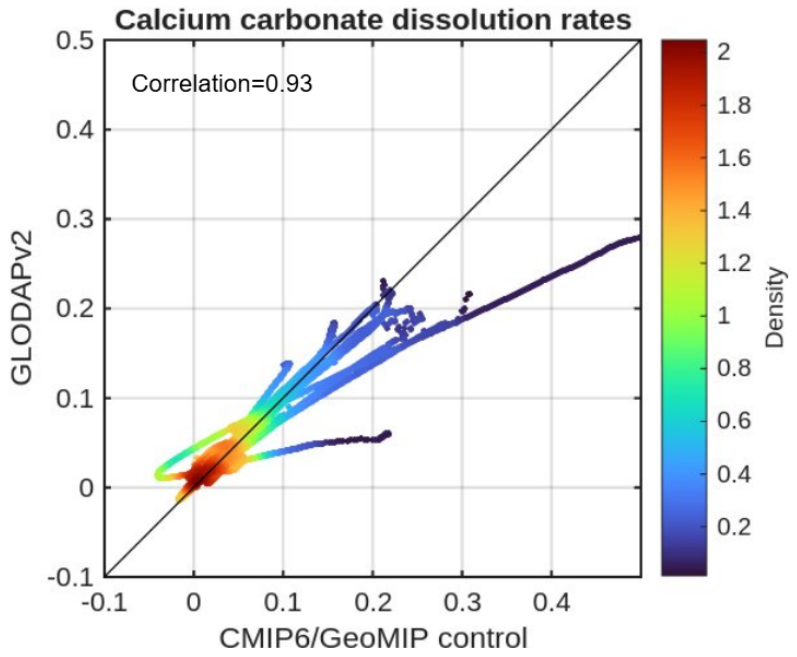

Figure 2: Shown, merging all biomes into one scatterplot, is a comparison of the calcium carbonate dissolution rate estimates using CMIP6/GeoMIP control simulation output and the GLODAP and other observationally-derived data used by Sulpis et al. (2021). The correlation of the quantities shown here is 0.93.

lationship. The influence of the other factors were not clear to me so I performed the comparisons as previously submitted. Because I made the mistake of using units of $molm^{-3}$ for oxygen, alkalinity, nitrate, phosphate, and DIC instead of the units they should have had ($\mu molkg^{-3}$), I performed the calculations again and found a surprisingly large sensitivity of the calcium carbonate dissolution rates to alkalinity in the GeoMIP output. The dissolution rates are actually most sensitive to mean age and next-most sensitive to alkalinity, not salinity. This means that the CMIP6 output-based analysis is an approximation because it assumes zero alkalinity change, whereas the GeoMIP output-based anal-

ysis is an approximation because it assumes only changes in mean age greater than 300 years. Near analysis is credible near the surface, particularly in subtropical regions and the subpolar North Atlantic Ocean, according to Wu et al. (2025) so excluding these regions (where waters tend to be younger than 200-300 years old), the GeoMIP-based analysis should be fairly accurate. An estimate of the alkalinity change for the CMIP6-based analysis is difficult to derive from the GeoMIP output, but we have attempted to do this using the relationship between salinity and alkalinity shown above. Estimates of the calcium carbonate dissolution rates using the CMIP6 output with these alkalinity changes are now shown in the manuscript as well.

- 5. Incomplete GeoMIP analysis Only 2 of the 6+ GeoMIP scenarios include Carbon-14 output to estimate mean age changes (line 76-77). Since mean age is identified as one of the two dominant factors, the comparisons of geoengineering effectiveness (e.g., Figure 7, conclusions) are incomplete and potentially misleading. The conclusions about which geoengineering techniques are "most effective" should be heavily caveated or removed.

- **Using Carbon-14 to get a mean age perturbation was included because you are right to suggest emphasizing the caveat that I do not have sufficient data to propertly characterize the mean age perturbation in the GeoMIP simulations. I tried to explain this caveat but will edit the text to more heavily stress this caveat. Here we show that there are differences in the CMIP6-based dissolution rate estimates when mean**

age changes in waters younger than 200-300 years old are included versus excluded, but these differences are insignificant. The magnitude of the differences are large in the subtropical North Pacific due to large uncertainties, but otherwise are small. The difference globally is 15.9%. Part of the purpose of the phase space figures is to show how sensitive the GeoMIP-based estimates are to the mean age. Additionally, we have attempted to account for the uncertainty in mean age in the GeoMIP calculations by propagating the estimated 15.9% bias in the final mean ages as an uncertainty.

- 6. Unquantified implications The manuscript repeatedly defers quantitative analysis: "Future studies will investigate the direct and indirect impacts of these changes more quantitatively" (line 298-299) "we don't know the full range of possible calcium carbonate dissolution rates" (line 290-291) "it is beyond the scope of the present study to quantify these effects" (line 249-250) If the implications cannot be quantified, the "so what?" question remains unanswered. What is the practical significance of a factor-of-two or order-of-magnitude change in dissolution rates? How does this translate to Pg C or mm of sea level?

- **I tried to quantify these numbers but could only arrive at very approximate numbers based on previous studies' findings and theories. For example, the back-of-the-envelope estimate of 84 mm of sea-level contraction over 150 years would become about 84 mm of additional sea-level contraction by 2100 (double the dissolution rate but half the time). But the amount of sea-level change will also depend on the spatial dis-**

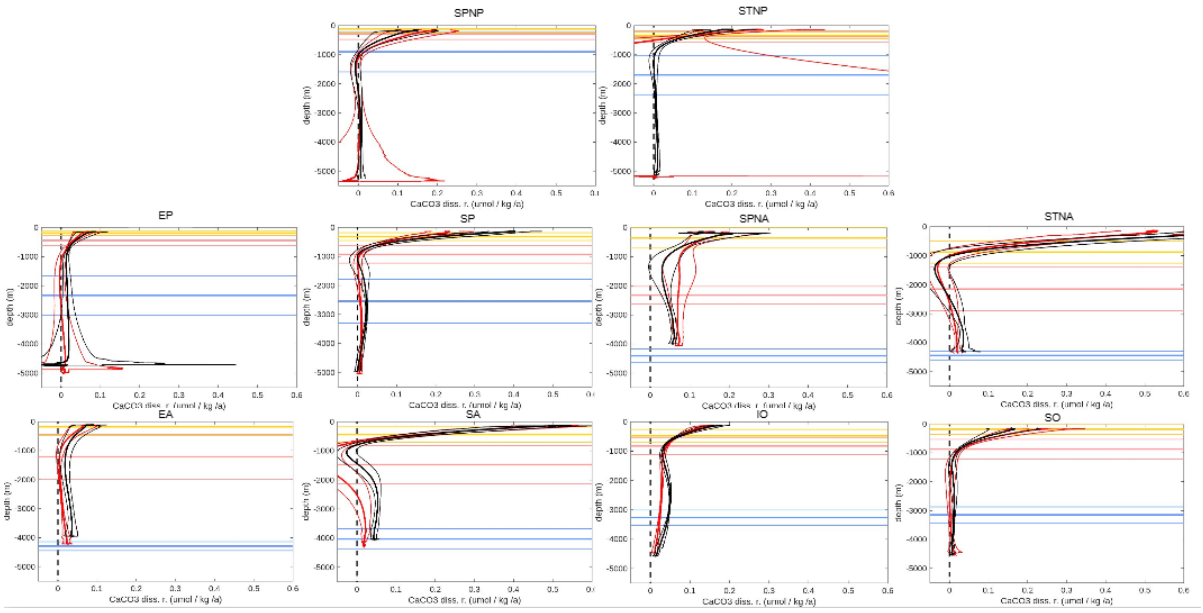

Figure 3: Shown for all ten biomes (SPNP - subpolar North Pacific, STNP - subtropical North Pacific, EP - equatorial Pacific, SP - South Pacific, SPNA - subpolar North Atlantic, STNA - subtropical North Atlantic, EA - equatorial Atlantic, SA - South Atlantic, IO - Indian Ocean, SO - Southern Ocean) are comparisons of the calcium carbonate dissolution rate estimates using CMIP6 output that include the influence of both the mean age perturbations on waters $<$ 200 years old and on Carbon-14-based age perturbations on waters $>$ 300 years old (black profiles) and the same, except the mean age perturbations on waters $<$ 200 years old is excluded (red profiles).

tribution of heat and freshwater in the ocean over the present-to-2100 time period, which in turn will also influence the carbon budget, and the carbon budget is difficult to predict because we don't know what the surface fluxes of carbon dioxide will be (let alone the surface fluxes of heat and salt) from our estimates. I would need to run a coupled sediment-ocean biogeochemically active future climate simulation in order to do a

more proper job. One purpose of this manuscript is to stress that these simulations need to be done because of how much the dissolution rate uncertainties propagate into sea-level/carbon budget uncertainties.

- 7. Section 4 – suspended sediments This section appears to be a separate mini-study that does not connect meaningfully to the main dissolution rate analysis. Specific issues: The assumption that TSM equals SSC and that all suspended matter is sediment (line 210-214) is crude. The connection between SSC in river deltas and open-ocean $CaCO_3$ dissolution is unclear. The "back-of-the-envelope" calculations (line 213) are insufficiently rigorous for a peer-reviewed manuscript. I recommend either removing this section or substantially expanding it with proper methodology and clearer connection to the main analysis.

- **This section has been removed from the manuscript.**

- 8. Figure quality and presentation Many results are described as "not shown" (e.g., lines 120, 130, 145). If these results are important enough to mention, they should be included in supplementary material. Figure 11 presents ranges across all biomes and scenarios but is difficult to interpret. Consider reorganizing or splitting into multiple panels. The biome numbering system (#1-#10) requires readers to constantly refer back to figure captions. Consider using abbreviated names instead.

- **The biome numbers have been replaced with basin abbreviations or removed altogether on the figure. I also now include figures in supplementary material instead of saying they are not shown.**

- 9. Writing clarity The manuscript is difficult to follow in places: The Methods

section (Section 2) is too brief and does not clearly explain how perturbations are applied. The transition between CMIP6 results (Section 3.1) and GeoMIP results (Section 3.2) is abrupt. The logic connecting dissolution rates → carbon budget → sea level is not clearly laid out.

- **Thank you for these suggestions. I have edited the text accordingly.**

- Technical Corrections 1. Line 217: Inconsistent minus sign format: "kg m$^{\smile}$3" should be "kg m^{3}" (appears multiple times) 2. Reference list: Check formatting consistency (e.g., some entries have issue numbers, others do not) 3. Line 20: "calcium dissolution rates" – missing "carbonate"? 4. Equation 2: The notation "ln(1)" equals zero, so this term appears unnecessary. Please clarify or simplify.

- **I have incorporated edits regarding the other specific suggestions, thanks.**