

## Review of Hinrichs et al. (2023)

### Overall comment

In overall, this work is interesting and innovative in its approach to bias assessment in OAE modelling studies using ESMs. Indeed, the study pushes a CMIP6 bias analysis (essentially focused on alkalinity, but also on DIC) towards a concrete, but idealized, CDR experiment. The approach used to decompose the alkalinity bias is broadly consistent with Planchat et al. (2023), thus reinforcing the message to the modelling community around the representation of alkalinity and the carbonate pump in ESMs. The writing of the manuscript could nevertheless be partly improved, in particular some parts could be restructured to gain clarity. Many technical corrections should be made.

### Specific comments

#### A. Main comments

1. In the decomposition of the alkalinity bias, it is repeatedly mentioned that “preformed alkalinity” refers to physical biases for the ocean, but it also contains biases of both organic matter and CaCO<sub>3</sub> production (e.g., l. 17 ; l. 121-125 ; l. 218 ; l. 291 ; l. 294)
2. The direct link made with the CO<sub>2</sub> uptake in the OAE idealized experiment is confusing for me (e.g., 274, l. 327): a relative  $p\text{CO}_2$  difference should not directly be a relative CO<sub>2</sub> uptake difference, or should it?

According to me, we have:

$$\frac{dDIC}{DIC} = \frac{1}{Re} \cdot \frac{dp\text{CO}_2}{p\text{CO}_2}$$

An ESM with a greater relative variation of  $p\text{CO}_2$  resulting from alkalinity addition, also has a greater Revelle factor. So, the product of the right member of the above equation should at least partly compensate. To access a first approximation of the CO<sub>2</sub> uptake associated with alkalinity addition, the relative variation of DIC should be expressed in function of the relative variation of alkalinity. I think that with what is currently offered, it is possible to assess the bias resulting from OAE in ESMs concerning acidification, but I’m confused by the direct link made with the CO<sub>2</sub> uptake.

#### B. Minor comments

The second part of the abstract could be improved and made more readable. In particular, I do not understand: “We find that the degree of compensation of DIC and alkalinity biases at the surface is more important for the marine CO<sub>2</sub> uptake capacity than the alkalinity biases themselves.” (l. 25-26); and l. 27-29 are too heavy, perhaps misplaced, and with a unit error (mmol/kg instead of mmol/m<sup>3</sup> as l. 209-211).

For the sake of clarity and consistency throughout the manuscript, I would suggest that alkalinity and DIC should always be shared in mmol/m<sup>3</sup> rather than mixing mmol/m<sup>3</sup> and umol/kg. It would simply be necessary to clarify how the conversions are done for GLODAPv2 and for using CO2SYS in the methodology (see l. 28-29, l. 149-152, Fig. 1 etc.).

Table 1: It would be great also to share the ensemble member for each ESM, rather than writing “the first available ensemble member” l. 109, which is slightly confusing.

The marine biogeochemical model of CNRM-ESM2-1 (not CNRM-ESM-2-1) is PISCESv2-gas. The one for IPSL-CM6A-LR is PISCESv2. The one for NorESM2-LM/MM is iHAMOCC.

Why do you precise the grid only for a few ocean models? If you wish to precise it, add it in the “ocean horizontal resolution” column and just precise maybe whether it is a tripolar grid or not.

I. 109-119: These lines would benefit from restructuring.

The TA\* method: are there limitations to this decomposition of the biases? What are the advantages of it rather than the one suggested in Sarmiento and Gruber (2006)? Not using DIC?

Eq. (3) and (4): why do you use phosphate for PO in Eq. (3) and then nitrate in Eq. (4) for TA\* rather phosphate (or nitrate) in both cases?

I. 139, you explicitly mention the fact that TA\* “is computed as residual after rearranging Eq. (2)”. This is questioning, since it is finally on the dissolution of CaCO<sub>3</sub> that we have the more certainty over its effects on alkalinity (+2 eq for 1 eq of CaCO<sub>3</sub> dissolved). On the contrary, the effect of remineralisation, for example, is more complicated to extract from ESMs because of the N-reactions and whether or not they are taken into account in the biogeochemical models. It is therefore perhaps a pity not to take advantage of the ease with which the carbonate pump can be used in terms of its effect on alkalinity with this method.

I. 156-157: Could you explain why you have chosen these values rather than the mean surface ESM values for instance?

I. 166-168: This sentence should be part of the methodology.

I. 190 and Fig. 3b: The very high TA<sub>n</sub> value observed at the ocean surface for GFDL-ESM4 is linked to very low salinity values (even possibly null) in the Baltic Sea if I remember well. You might be able to neglect this closed sea for this ESM when averaging to avoid this issue. You could also use a salinity threshold maybe.

Fig. 3 and 4: I would combine both figures so that the first panel is not repeated.

I. 201: It could be mentioned that the bias observed for CNRM, IPSL and UK is associated with the ocean model NEMO.

I. 211: “TA biases likely lead DIC biases, as DIC can adjust through gas-exchange of CO<sub>2</sub>”. I suggest to add at the end “to maintain a surface chemical equilibrium with the atmospheric CO<sub>2</sub> concentration”. Besides, this aspect could be slightly more highlighted to understand why you decompose the alkalinity biases and not the DIC biases in addition, whereas you consider both surface alkalinity and DIC in the following with CO2SYS.

I. 221-230: This paragraph should be rewritten.

It should be readable and meaningful independently of Fig. 6 (avoiding “are shown in Figure 6” for instance).

The first sentences are too repetitive compared to what was already described in the previous sections.

I. 226-228: This sentence is out of topic as you are talking about biases in this section.

It would be great also to share some values (in absolute and/or %).

Fig. 6: The addition of MMM would be meaningful on these plots.

I. 237: I am not convinced of the veracity of the causal sequence (cf. “thus”). Could you detail it?

I. 240-242: “All panels are sorted by Revelle factor in ascending order.” This is enough in the legend. “The Revelle factor ... at the ocean surface.” should be part of the Introduction.

Fig. 7: Could you set “MMM” in bold so that we can easily spot it?

I also suggest to keep the x-axis increasing in the last panel, to avoid confusions.

I. 260-261: Too repetitive with what was already said for Fig. 1.

I. 262: It is less than 13.0 % in Fig. 8, I think: t should be about 10.7 %.

I. 263: It would be great to have also the TA bias in Fig. 7 with the ESMs ordered in the same way.

Fig. 8: Could you set “MMM” in bold so that we can easily spot it?

Could you adapt the y-axis scale to avoid the blank space to the top?

Discussion and conclusions: In overall, this section could be improved in terms of organization and content. There are some repetitions and it is sometimes difficult to follow you.

In particular, l. 310-323 are quite messy for instance and the points overlap each other sometimes. In the first point, are you pointing towards including aragonite as well as calcite? What would be the effect of reducing the calcification rate in the Southern Ocean, where deep waters are upwelled? l. 320: are you talking about calcite dissolution (effectively sometimes explicitly modelled) or calcite production (always implicitly modelled in that case)? Another important point, which is not mentioned, is the burial and dissolution at the seafloor.

Fig. S2: It is not compared to the GLODAP climatology here, as opposed to Fig. 6.

Table S1: Not useful, I think.

There might be some issues in the References. There is at least one for Planchat et al., 2023, since some authors are missing.

### Technical corrections

*The following remarks are valid for the whole manuscript, and only a few occurrences will be mentioned below:*

- There is a regular lack of punctuation, mainly commas.
- Beware of citations: a number of references appear with double brackets in the text.
- In intercomparison studies of ESMs, it may be more accurate and preferable to refer to ESMs throughout the study rather than models.
- If you decide not to put a space before '%', do so everywhere.
- I suggest to write 'TA\* method' instead of 'TA\* Method'
- The p in  $p\text{CO}_2$  should be in italic
- When the abbreviation/acronym has been defined, it can then be used directly (especially in the legend of the figures)
- Try to keep the text independent of the figures and mention the figures in parentheses instead of within the text directly
- Write "Revelle factor" instead of "Revelle Factor"
- Always write "TA-to-DIC-ratio" instead of mixing the way you mention it

l. 4: 1 should in exponent

l. 9-15: commas are missing

l. 10: ... on alkalinity ...

l. 10: That is why, in the search for ...

l. 11: ... Ocean Alkalinity Enhancement (OAE) ...

l. 12: ... exists on how ...

l. 13: ... 14 CMIP6 Earth system models (ESMs) ...

l. 17: ... shows ...

l. 36: ... overestimate it in the ...

l. 40-42: commas are missing

l. 45: ... species composing the so-called ...

I. 45-47: Suggestion: The oceanic uptake of anthropogenic carbon leads to an increase in aqueous CO<sub>2</sub> and thus DIC. By changing the chemical equilibria between the carbonate species, this results in ocean acidification with a decrease in pH.

I. 48: ... over acids (proton donors) ...

I. 48: ... role in the partitioning of the DIC pool, especially in the form ...

I. 49-56: Very nice paragraph, but the first sentence could potentially be improved. Suggestion: The carbonate system is key in driving the seawater ability to resist a change in its chemistry, also called its buffering capacity. In particular, the Revelle factor is the sensitivity of pCO<sub>2</sub> to changes in DIC. A low Revelle factor indicating a high buffering capacity and vice versa.

I. 55: ... the potential CO<sub>2</sub> uptake ...

I. 68: ... much research has focused on ...

I. 72: ... (OAE; Köhler et al., 2013 ...

I. 81: ... set-ups (e.g., Ilyina et al., 2013; ... ; Burt et al., 2021).

I. 86-87: ... underway, or in planning, seeking to apply ... for OAE (e.g., Butenschön et al., 2021), highlighting the importance and urgency of a robust ESM evaluation.

I. 93: They report an improvement in ...

I. 109: "the first available ensemble member" is confusing and not precise.

I.111: (CDO; Schulzweida, 2022)

Eq. (1): Precise that S is the salinity

I. 115-119: Suggestion: ... against gridded observational products: (i) TA, DIC and pH from the GLODAPv2.2016b Mapped Climatology (GLODAP in the following; Lauvset et al., 2016); (ii) oxygen and nutrients from the World Ocean Atlas 2018 dataset (WOA; Garcia H.E., 2019) and GLODAP; and (iii) salinity and temperature from the Polar science center Hydrographic Climatology (PHC3.0; Steele et al., 2001) and WOA. For the evaluation of global mean vertical profiles, the model data are interpolated onto the same 33 vertical levels used in the GLODAP climatology.

I. 125: ... calcium carbonate (CaCO<sub>3</sub>) dissolution (TA\*)

I. 128 : potential temperature

I. 129 : if NO is mentioned, then it should be defined to explicitly mention the difference with PO.

I. 131, 136: could you add references for the ratios r\_(...) that you consider?

I. 131-132: "onto upper ocean TA values" and then "interior ocean" is a bit contradictory

I. 140: ... to 10 of the 14 CMIP6 ESMs (... NorESM2-MM and UKESM1-0-LL)

I. 143: 2.3. Theoretical model sensitivity to OAE

I. 145: OAE. Thus, ...

I. 147-148: Suggestion: this toolbox, from the combination of two of the carbonate system variables, compute the entire ocean CO<sub>2</sub> system.

I. 149-152: Suggestion: ... 1,026 kg/m<sup>3</sup> (Fig. 1). First, we evaluate the CO<sub>2</sub>SYS output fields : Revelle factor, pH and  $p\text{CO}_2$  (partial pressure of CO<sub>2</sub> in seawater) based on the CMIP6 outputs. Then, we assess the changes...

I. 155 ... (MMM). Black vertical ... value. (b) Same ...

I. 156: potential temperature

I. 164: ... at the surface (Fig. 1a and 2)

I. 165: (Fig. 1a and 2)

I. 170: ... in NorESM-LM/MM and CanESM5; Fig. S1).

Fig. 2: Panel 1, title: GLODAP

Panel 2, title: GLODAP error; colorbar title: absolute error.

General colorbar title: Surface TA bias [mmol/m<sup>3</sup>]

Legend: Surface distribution of TA in GLODAP (top left), its error estimate (top center) and the CMIP6 multi-model-mean (MMM) bias (top right), as well as the respective biases of the ESMs.

Same comments for Fig. S1, where TA<sub>n</sub> should be directly mentioned.

Same comments for Fig. 5.

Try to homogenize the legends too

I. 177: ... the increase of TA with depth deeper ...

I. 179: ... TA overall. This indicates that their global inventory of TA is respectively too low and too high ...

I. 182: ... in the GFDL ESMs ...

I. 185-186 1,024; 1,028; 1,035

I. 187: ... at depth, or vice versa, ...

I. 189: ... in the upper ocean. The surface minima ...

Fig. 3: Could the black and grey lines have no transparency and be to the forefront so that we can clearly see both of them?

Legend: ... and TA<sub>n</sub>, (b) ...

I. 199: ... by the ESMs, referring to circulation biases.

I. 200: ... Ocean), between ... depth, ...

I. 208: ... and local distribution ...

I. 216: 3.2 Decomposition of the vertical alkalinity biases

I. 219: ... (CaCO<sub>3</sub>) dissolution and ...

Fig. 6: subplot titles: I suggest to simply write "TA/TA<sup>0</sup>/TA<sup>r</sup>/TA\* bias"

I. 235: Impact of biases on OAE efficiency

I. 236: ... surface TA and DIC ...

I. 238: ... was conducted, using surface TA and DIC, to calculate the full carbonate system (see Methods and Fig. 1).

I. 248: ... GLODAP data, ranging from ...

Fig. 7: ... CO2SYS toolbox. The results ...

I. 269: ... OAE idealized experiment shows that 12 out of 14 ESMs ...

I. 278-279: ... mmol/m<sup>3</sup>; i.e., YY %)

I. 294-295: In the sub-surface and the deep ocean, biases in TA are also driven by the CaCO<sub>3</sub> dissolution, while contributions from remineralization of organic matter are negligible.