

Foreword

Both reviewers have not fully perceived the message of my work. Therefore, first I give a short overview of the basics of my paper. I wanted to provide a better understanding of the basic operation of the physical pump. This pump works in two steps. First, the carbonate buffer system of the upmost mixed layer reacts with the CO₂ of the atmosphere to attain chemical equilibrium. The question is: what is the increase of DIC in the mixed layer when the partial pressure of CO₂ in ppm increases by ΔCO_2 . To this end I have calculated DIC (mmol/kg) as a function of CO₂ (ppm) using the program PHREEQC. Differentiating with respect to CO₂ gives $d\text{DIC}/d\text{CO}_2$ (mmol/kg ppm) termed as sensitivity S as a function of CO₂. S is the increase of DIC by increase of CO₂. It tells how much CO₂ is absorbed by the buffer system. S decreases with increasing CO₂. Usually, this buffering is described by the Revelle factor R . Therefore, I had to give the relation between S and R . Although, this is a side result it requires some text. Both reviewers have focussed to this part of the paper. The essential second part of the pump is transport of water of the mixed layer with high DIC by thermo-haline circulation into deep ocean and replacement by water in equilibrium with preindustrial CO₂ level. This part of the pump increases steadily with increasing CO₂. I admit that this is a simple model that needs only the well-known constants of equilibrium chemistry provided by PHREEQC and the amount of waterflow into deep ocean in S_v . The result gives at least the correct order of magnitude of the observed CO₂ uptake from the atmosphere into the ocean by the physical pump. In summary, my model reveals the basics that may be hidden in many complex models that are not intelligible by non- specialists. This opens understanding to a larger part of the scientific community and to my knowledge has not been published before.

Reply to RC2:

In the following the arguments of the reviewer are in *italic*, my response is in normal, and parts copied from my paper are underlined.

There is some misunderstanding in the definition of the Revelle factor. The reviewer's definition is

$R = (\text{DIC}/[\text{CO}_2]) / (\Delta\text{DIC}/\Delta\text{CO}_2) = (\Delta[\text{CO}_2]/[\text{CO}_2]) / (\Delta\text{DIC}/\text{DIC})$ where $[\text{CO}_2]$ is the aquatic CO₂ concentration; both DIC and $[\text{CO}_2]$ are measured in gravimetric units (mol kg⁻¹).

My definition is $R = (\Delta\text{DIC}/\text{DIC})/(\Delta\text{CO}_2/\text{CO}_2) = (\Delta\text{DIC}/\Delta\text{CO}_2)/(\text{DIC}/\text{CO}_2)$. ΔDIC is the change in concentration DIC caused by a small increase $\Delta\text{CO}_2(\text{gas})$ of the concentration CO₂ in the atmosphere. ΔDIC and DIC is in mol/kg and) ΔCO_2 and CO₂ is in ppm, the partial pressure of CO₂(gas) in the atmosphere. Since the aquatic CO₂ concentration CO₂(aq) is related to the partial pressure CO₂(gas) in the atmosphere by Henry's law CO₂(aq) = $K_H \cdot \text{CO}_2(\text{gas})$ and $\Delta\text{CO}_2(\text{aq}) = K_H \cdot \Delta\text{CO}_2(\text{gas})$ both definitions are identical.

By rearranging one gets $R = (\Delta[\text{CO}_2]/[\text{CO}_2]) / (\Delta\text{DIC}/\text{DIC}) = (\text{DIC}/\text{CO}_2)/(\Delta\text{DIC}/(\Delta\text{CO}_2)) = (\text{DIC}/\text{CO}_2)/S$ for both definitions and switching units in the lengthy comment is not necessary.

In line 160 ff in my work on finds: From its definition a simple relation is: $R = 2.27/(\text{CO}_2 \cdot S)$ because $\text{DIC} \approx 2.27 \text{ mmol/L}$ remains constant within a few percent (see Fig. 1 and Fig. 5).

After this lengthy discussion of relations between R and S, RC2 writes:

"This is a misunderstanding, because with increasing atmospheric CO₂ the uptake of CO₂ by the ocean and its transport to deeper layers (solubility pump) will further increase. This is consistent with a decrease of the buffer capacity of the ocean with respect to increasing atmospheric CO₂ which can be expressed by an increase of the Revelle factor or a decrease of the sensitivity (inversely related to each other). Based on this misunderstanding, the author addresses a problem that does not exist. I can not support publication of this paper."

To resolve this misunderstanding I will add in the revision: $R = (\Delta\text{DIC}/\text{DIC})/(\Delta\text{CO}_2/\text{CO}_2) = (\Delta\text{DIC}/\Delta\text{CO}_2)/(\text{DIC}/\text{CO}_2)$. ΔDIC (mol/L) is the change in concentration DIC (mol/L) caused by a small increase ΔCO_2 (ppm) of the concentration CO_2 (ppm) in the atmosphere. CO_2 can be also given as the concentration of aqueous CO_2 in mol/L because $\text{CO}_2(\text{atm})$ and $\text{CO}_2(\text{aq})$ are related by Henry's law; $\text{CO}_2(\text{aq}) = K_H \cdot \text{CO}_2(\text{atm})$.

What is the misunderstanding? Such general remarks are not helpful.

Evidently, the reviewer has missed the second part of my paper where transport into the ocean is discussed. See Fig. 11 and Fig.5 and lines 249-257.

The total solubility sink consists of two mechanisms: The equilibrium pump as described and the transport pump that is caused by the global meridional overturning circulation of 36 Sv.

This transfers into deep-ocean the difference $(\text{DIC ppm} - \text{DIC } 280) = \Delta\text{DIC ocean}$ that has been accumulated in the mixed layer from onset of industrialisation to the actual CO₂ level.

This sink increases continuously replacing the failure of the equilibrium pump. At 400 ppm the total sink is 1.9, at 600 ppm it is 3.8 and at 800 ppm it amounts to 5 Gtons/year depending solely on the CO₂ level in the atmosphere. For ppm > 600. To conclude, the total solubility pump is not endangered by ocean acidification. In contrast, it increases with increasing CO₂ level of the atmosphere to yield significant contribution to remove anthropogenic CO₂ from the atmosphere into deep-ocean.

Detailed remarks to RC2:

p.2 'At 400 ppm a value of about 1.9 Gtons/year is estimated that increases to 3.9 Gtons/year at 600 ppm and to 5 Gtons/year at 800 ppm.' Sentence needs more explanation ... 1.9 Gt C oceanic net uptake; estimated in current paper or from literature?)

I do not understand this. Evidently this is estimated in the current paper.

“Units: Gt C or Gt CO₂? I guess always Gt C”

units are in Gt CO₂. See line 169-170. The numbers are derived in the current paper, see Fig. 11. I will use Gt CO₂ instead of Gt throughout the text, (mol/(Lppm))

p.4 The input and output table of PHREEQC should be replaced by a proper list of relevant quantities with appropriate units (for example: what is meant by ‘CO₂(ag) - 2.921’) The sensitivity as a function of DIC, TA, temperature, and salinity is easy to calculate with freely available software packages (compare, for example, Orr et al., 2015, Orr & Epitalon, 2015, Humphreys, et al, 2022, especially with CO₂SYs available in MATLAB or Python on GitHub: <https://github.com/jamesorr/CO2SYS-MATLAB>, <https://github.com/mvdh7/PyCO2SYS>).

I used the input and output files of PREEQC. All units are explained in the text. Maybe other programmes do the same job. But with the information given the reader can do calculations By PHREEQC.

In the following I comment some of the special remarks.

p.6 ‘which 9 units have reacted to carbonates. For low pH < 4 where all DIC is in CO₂aq, S = 1. At 15°C the value of S = 0.0001 corresponds to value S* = 2.5. ’ S, S*:
units missing.*

From 10 CO₂ molecules absorbed 9 are converted to carbonates. Units of S are defined in the text.

p.10 ‘Therefore, dDIC/dCO₂ = KH’ is wrong!

The statement is correct only for pH < 4 when only CO₂(aq) is existing. It was discussed to provide the limit at low pH.

p.9 ‘From this one may understand why R is used only qualitatively [TYPO] to judge ocean’s physical pump buffer capacity.’ ???

There are many comments with ????? but their question remains open.

I am helpless how to react to this review. I am afraid that the reviewer had only a restricted perception of the paper to provide a constructive review.

In conclusion the review is highly biased to decline the paper and most of the objections are unfounded or even wrong. It does not give any hints how to improve the paper. Therefore, I cannot suggest any changes to the paper at present. I leave it to the editor how to proceed.

