Seasonal dynamics and annual budget of dissolved inorganic carbon in the northwestern Mediterranean deep convection region

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Responses to the comments of the anonymous Reviewer 2

First we would like to warmly thank Reviewer 2 for his/her relevant and constructive comments which will help to improve the manuscript.

Answers to reviewers' comments are reported point by point. The questions and comments of the anonymous Reviewer 2 are in black, the responses in blue and the modifications that we propose for the revised manuscript in red in italic.

Review of Ulses et al. (2022): "Seasonal dynamics and annual budget of dissolved inorganic carbon in the northwestern Mediterranean deep convection region"

Summary This study by Ulses and coauthors presents a detailed carbon budget in the deep convection area of the NW Mediterranean Sea for the period between September 2012 and September 2013. Using an ocean biogeochemistry model forced with daily output from a physical model, the authors find that their focus region is a moderate sink of atmospheric CO2 over the study period. In addition, by dividing the study area into the upper 150m and the deep ocean below that, they find that both physical and biological fluxes play an important role in controlling carbon fluxes across seasons.

Overall, the authors did a great job comparing their model results to existing observations and presenting the carbon budget of their study region in great detail. Therefore, the study is generally suitable for publication in Biogeosciences. However, I would like to raise several points, mostly regarding the presentation of the study, which should be addressed before the publication of this manuscript.

Please see the detailed explanation of these major points and all my detailed comments below.

We appreciate this positive general assessment.

Main comments

1. Introduction: Acknowledging that I am not 100% familiar with the literature concerning the Mediterranean Sea, the introduction appears to give a good summary of previous work. However, it fails to make the knowledge gap clear enough in my view. In its current form, it still reads too much as a collection of results from individual studies, making it hard for the reader to figure out what has not been addressed or what the short-comings in each of these previous studies are. As a result, I am a bit lost guessing what exactly the focus of the study by Ulses et al. is until L. 108. I suggest revising the introduction to more clearly state where the knowledge gaps are that are to be addressed in this new study.

Response: As suggested by Reviewer 2, we will revise the introduction to more clearly point the gaps in the previous studies on DIC cycle and give earlier in the text the objective of the present study, as follows:

"Northern deep regions of the semi-enclosed Mediterranean Sea, i.e. the northwestern region (Fig. 1, Gulf of Lion and Ligurian Sea) and the South Adriatic, located at mid-latitudes are ones of the regions where deep convection occurs (Ovchinnikov et al., 1985; Mertens and Schott, 1998; Manca and Bregant, 1998; Gačić et al., 2000; Béthoux et al., 2002). Few studies have investigated the dynamics of dissolved inorganic carbon (DIC hereafter) in the northwestern Mediterranean Sea, where the Western Mediterranean Deep Water is formed and which plays a crucial role in the circulation and ventilation of the Mediterranean Sea (Schroeder et al., 2016; Li and Tanhua, 2020; Mavropoulou et al., 2020). The objective of this study is to gain insights on the annual cycle of DIC by examining and quantifying the biogeochemical, physical and air-sea fluxes.

In the northwestern Mediterranean region, a basin-scale cyclonic gyre is associated with a doming of isopycnals. The density increase, induced in winter in surface waters by cold and dry northerly winds, produces instabilities of the water column leading to convective mixing of surface waters with deeper waters. With regards to the biogeochemical processes, the region is characterized at the sea surface by a moderate phytoplankton bloom in fall, interrupted by deep winter mixing, and an abrupt phytoplankton bloom, following deep winter mixing which has supplied inorganic nutrients to the euphotic layer (Severin et al., 2014; Bernardello et al., 2012; Lavigne et al., 2013; Ulses et al., 2016; Kessouri et al., 2017). At the annual scale, the net community production (NCP, defined as the gross primary production minus the community respiration) was found positive leading to an autotrophic status of the area (Ulses et al., 2016; Coppola et al., 2018). The downward export of organic carbon and its interannual variability have been related to the intensity of the deep convection and the bloom (Heimbürger et al., 2013; Herrmann et al., 2013; Ulses et al., 2016).

Previous observational and modeling studies that have documented the dynamics of the CO_2 system in this region mostly focused on the Ligurian Sea, at fixed sites (Hood and Merlivat, 2001; Copin-Montégut and Bégovic, 2002; Bégovic and Copin-Montégut, 2002; Mémery et al. 2002 ; Copin-Montégut et al., 2004; Touratier and Goyet, 2009; Merlivat et al., 2018; Coppola et al., 2020), where the intensity of convection generally remains moderate compared to the Gulf of Lion. These 1D studies showed a pronounced seasonal cycle of pCO_2, mostly controlled by the sea surface temperature. The thermal effect is counterbalanced in spring by the impact of phytoplankton growth which leads to DIC drawdown, and in winter, by intense mixing events which bring DIC rich-water to the surface (Hood and Merlivat, 2001; Mémery et al. 2002 ; Copin-Montégut et al., 2004). On an *annual timescale, the Ligurian Sea was found to be a medium to minor sink for atmospheric CO_2 (Hood and Merlivat, 2001; Mémery et al. 2002 ; Copin-Montégut et al., 2004; Merlivat et al., 2008). Based on data, Touratier et al. (2016) complemented those observations from the Ligurian mooring sites, by describing the distribution of the carbonate system properties in the central region of the deep convection region during two winter periods, during and just after the deep convection event. The authors showed a rapid transfer of anthropogenic CO_2 to the ocean interior during the convection event and reported an excess in CO_2 in surface waters related to the atmosphere. Finally, D'Ortenzio et al. (2008) and Cossarini et al. (2021) based on a 1D model and a 3D model, respectively, found that the whole deep convection region is a major sink of atmospheric CO_2 in the open Mediterranean Sea. In the previous studies, the 3D dynamics of the CO_2 system over an annual cycle has never been specifically explored for the whole northwestern Mediterranean convection region and a complete DIC budget is still lacking for this region."*

2. Description of the model setup: While being methodologically sound from what I understood, the description of the model setup in section 2.1.2 is currently hard to follow. I suggest including a sketch in the revised version of the manuscript illustrating the downscaling approach and providing information on the initialization and run time of the simulations in each of the steps of the setup.

Response: We will clarify this description by adding a figure in Supplementary Material to show (1) the domain of the two coupled physical-biogeochemical models, i.e. the parent and child models, and (2) a scheme of the dowscalling strategy.

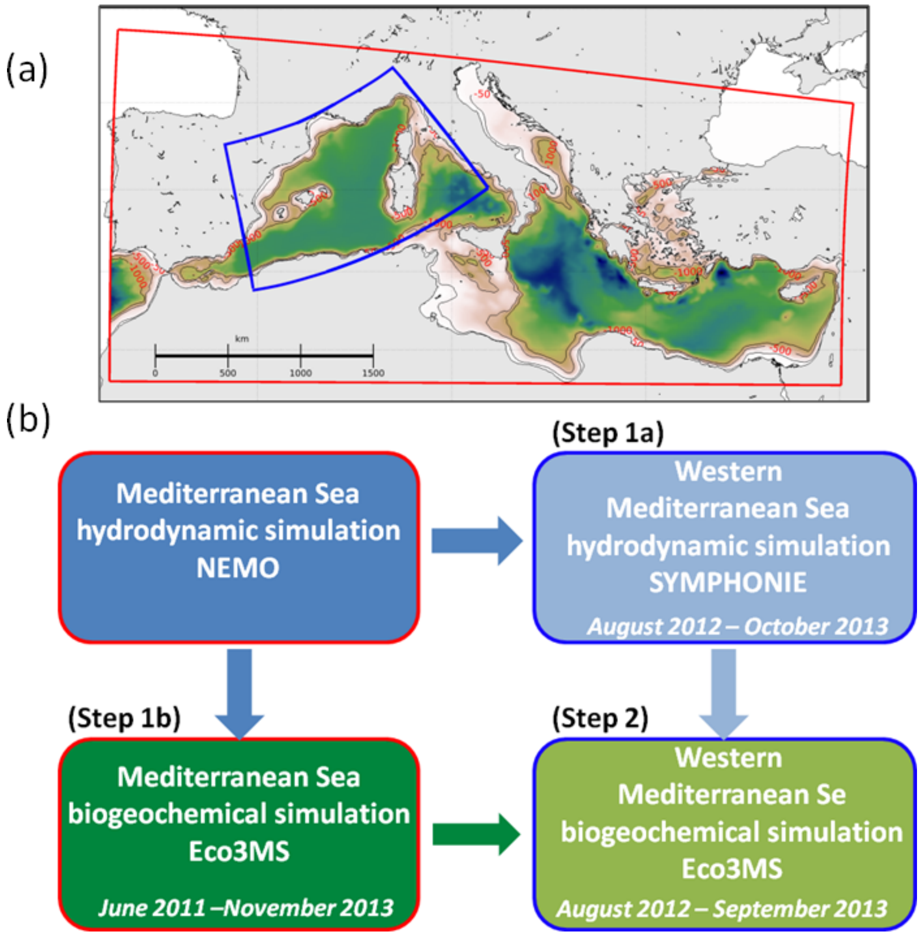


Figure S1: (a) Domain and bathymetry (m) of the forcing coupled NEMO-Eco3MS Mediterranean model (red contour) and of the coupled SYMPHONIE-Eco3MS western sub-basin model (blue contour). (b) Scheme of the downscaling strategy from the Mediterranean Sea to the western sub-basin.

We will also add in Section 2.1.3 "Model setup" of the revised manuscript the run time of each of the three simulations and will move the description of the initialization in step 1b before describing step 2. We hope the description of the downscaling strategy will be clearer after these additional elements and modifications:

"The implementation of the hydrodynamic simulation and the strategy of downscaling from the Mediterranean Basin to the western sub-basin scale in three stages have been described in detail in Estournel et al. (2016) and Kessouri et al (2017) and will be summarized here (Fig. S1):

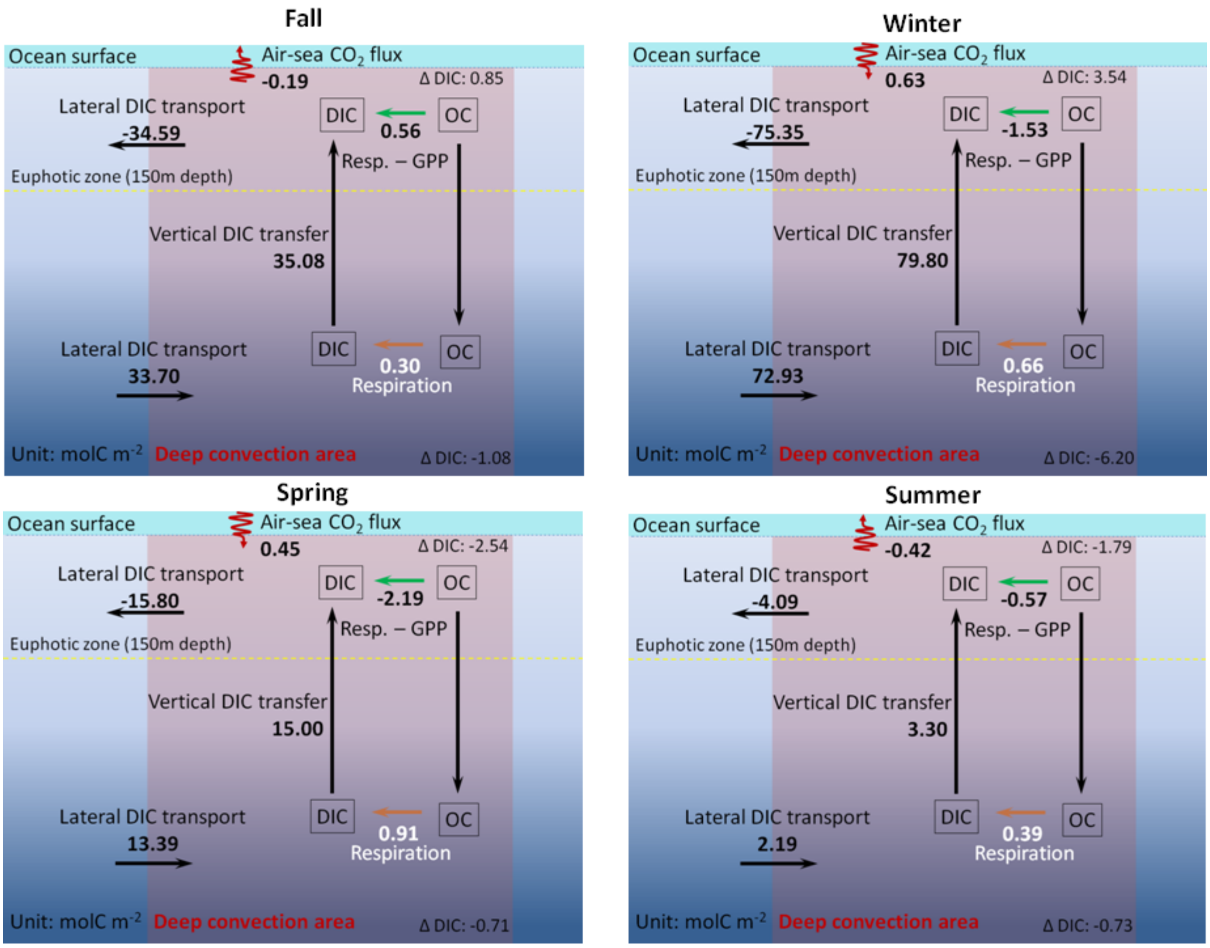
- In a first step (named step 1a), the SYMPHONIE hydrodynamic model, implemented over the Western Mediterranean Sea (delimited by blue lines in the insert of Fig. 1), was initialized and forced at its lateral boundaries with daily hydrodynamic analyses of the configuration PSY2V4R4, based on the NEMO ocean model at a resolution of 1/12° over the Mediterranean Sea by the Mercator Ocean International operational system (Lellouche et al., 2013). This simulation was performed from 1st August 2012 to 31 October 2013.

- In parallel (step 1b), the biogeochemical model was computed, in offline mode, at the Mediterranean basin scale, on the same 1/12° NEMO grid (delimited by orange lines in the insert of Fig. 1), using the same NEMO hydrodynamic fields as those used by the SYMPHONIE simulation in step 1a. This simulation was performed from 15 June 2011 to 15 November 2013. The carbonate system module in this configuration was initialized using mean values of dissolved inorganic carbon, total alkalinity observations carried out in 2011 from the Meteor M84/3 (Alvarez et al., 2014), CASCADE (CAscading, Surge, Convection, Advection and Downwelling Events, Touratier et al., 2016), and MOOSE-GE cruises (Testor et al., 2010) and at the EMSO-DYFAMED mooring (Coppola et al., 2021) and BOUSSOLE buoy (Golbol et al., 2020) sites, over bio-regions defined in Kessouri (2015), based on Lavezza et al. (2011). We deduced the concentration of the excess negative charge based on nutrient concentrations initialized using the Medar/Medatlas database as in Kessouri et al. (2017). Recently, Davis and Goyet (2021) described a method based upon the property variability, to precisely quantify the uncertainties at any point of an interpolated data field. This approach could be used in the near-future to improve both the at-sea sampling strategy (Guglielmi et al., 2022a; 2022b), and the accuracy of model initialization.

- In a second time (step 2), the Eco3M-S biogeochemical model was implemented over the Western Mediterranean Sea, using the grid and the hydrodynamics fields of the aforementioned SYMPHONIE simulation (step 1a) in offline mode. This simulation was performed from 15 August 2012 to 30 September 2013. The initial state and lateral boundary conditions of the biogeochemical fields are provided by the biogeochemical simulation of the Mediterranean Basin of step 1b. "

<u>3. Result section 4.1</u>: I admittedly found it quite difficult to keep up with all the provided details in this section. In general, I appreciate the detailed description of the figures, and I generally think the clear division into the different seasons is good. However, this division means that the reader must constantly jump back and forth between Fig. 6-11, making it very important to have consistent structure and summarizing sentences throughout this section. While such summarizing sentences already exist for some of the seasons (see e.g., winter sub-period 2), they do not for others. I thus suggest that the authors carefully screen the result section again to structure the description of each season as consistently as possible and that they add clear summarizing sentences to each of the seasons. I encourage the authors to work on the paragraph structure (including topic sentences), as this will greatly improve the readability of this part of the manuscript. Lastly, since I really appreciated Fig. 12 as a summary for the annual mean budget, a similar figure for the seasonal budgets (=1 figure, 4 panels) would be a valuable addition to the paper and would serve as guidance for the reader throughout section 4.1.

Response: In the revised manuscript, we will structure the description of the different seasonal sections as consistently as possible, with a description of the (1) atmospheric and hydrodynamical situation, then of the (2) biogeochemical fluxes, (3) physical fluxes, (4) air-sea fluxes, and (5) finally of the resulting variation of DIC content, and a summary of the budget in the upper layer. Besides, we will include in Figure 7 a panel with a similar figure as Figure 12 for each season, and remove from Figure 7 the panel (b) to avoid repetitions with the new sub-figure. We will merge Figures 8 and 9 to decrease the number of figures in this part.



"Figure 7c: Scheme of cumulative seasonal fluxes in mol C m^{-2} over the respective periods (fall: 88 days, winter: 116 days, spring : 74 days and summer: 87 days). Resp. stands for respiration and GPP for gross primary production."

<u>4. For the sensitivity experiment regarding calcification:</u> I was surprised to see an enhanced oceanic CO2 uptake relative to the reference case in the experiment accounting for calcification. For such an experiment, I would expect less oceanic CO2 uptake, given that the impact of calcification on alkalinity is twice that on DIC (thus increasing seawater pCO2 at the surface). Going back to your method section 2.1.4, I noticed that you only specified the impact of calcification on DIC – did you also include its impact on alkalinity in your sensitivity test? How did you parametrize dissolution at depth? I note that I realize that either way, this

will not impact the outcomes of the main findings of this study, but if this was indeed a mistake, I suggest that the authors correct it.

Response: We thank Reviewer 2 for raising this point. We acknowledge there was an error in the sensitivity test on calcification process, by omitting to take into account the process in the rate of change of alkalinity (excess negative charge denoted $\Sigma[-]$). We apologize for this error. We have corrected it by adding in the equation of the rate of change of alkalinity (excess negative charge denoted $\Sigma[-]$) the term of calcium carbonate production added in the DIC equation multiplied by 2 (Middelburg et al., 2019). In the new results, the air-sea flux could be reduced by 16% to 57% in considering calcification processes. We will modify the text and Figure 14 in the discussion section on the sensitivity tests on air-sea CO_2 flux, in Sect. 2.1.4 "Sensitivity tests" and in the conclusion. The dissolution at depth was not taken into account in these sensitivity tests.

Middelburg, J. J.: Marine Carbon Biogeochemistry A Primer for Earth System Scientists, Springer B., edited by Springer Briefs in Earth System Sciences, Springer Briefs in Earth System Sciences, 2019.

Section 2.1.4 Sensitivity tests :

"Thus, if we assume the ratio of calcium carbonate production to NCP is close to PIC:TOC, we added in Eq. 1 a consumption term representing 36% (for the mean value of PIC:POC ratio, 22% and 58% for the minimum and maximum ratio values, respectively) of NCP. This term, multiplied by 2, was added in the equation of the rate of change of the excess negative charge."

Discussion section:

"Finally, sensitivity tests taking into account supplementary consumption terms in the equation of DIC and excess of negative charge for $CaCO_3$ precipitation (Sect. 2.1.4) were performed to assess its potential influence on air-sea CO_2 flux. They show that not taken into account calcification processes could lead to an ~~underestimation~~ overestimation of the annual air-sea CO_2 uptake by ~~23~~16 to ~~58~~57% with estimates ~~of 0.72~~0.29 mol C m^{-2} yr^{-1}, based on the mean PIC:POC ratio given by Miquel et al. (2011) (varying between 0.20 and 0.36 mol C m^{-2} yr^{-1} based on the maximum and minimum PIC:POC ratios, respectively), and ~~0.58~~0.40 mol C m^{-2} yr^{-1}, based on the parametrization used in Lajaunie-Salla et al. (2021)."

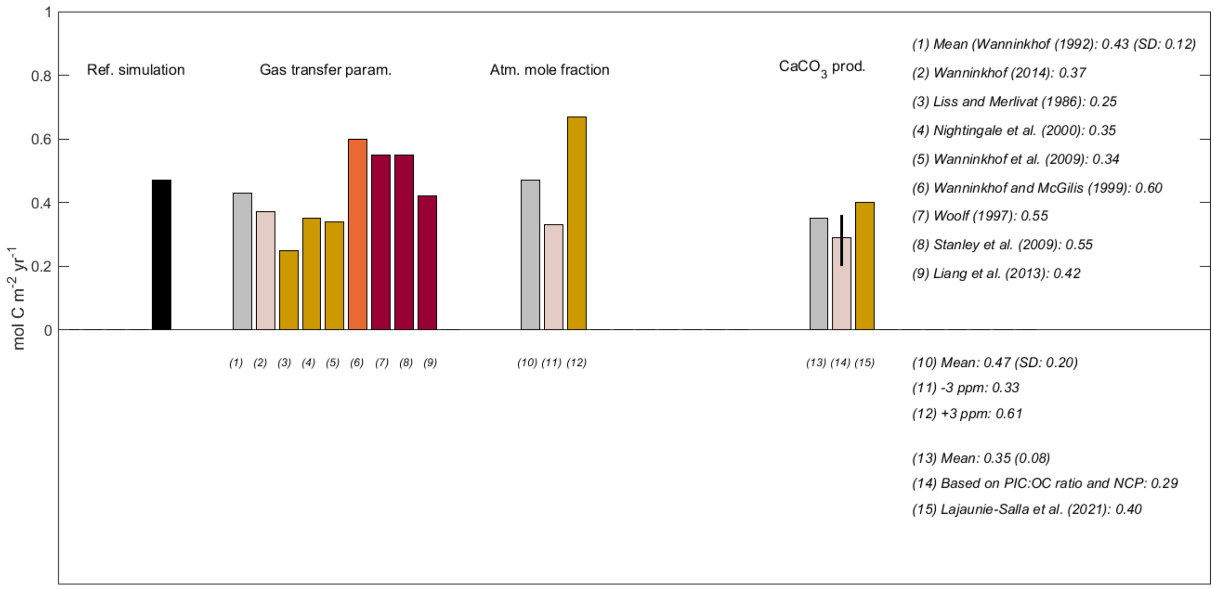


Figure 14: Sensitivity tests to the parameterization of gas transfer velocity, the variability of the mole fraction of CO_2 in the atmosphere, and the calcification processes on the annual CO_2 air-sea flux estimate. The black bar indicates the annual estimate in the reference simulation, grey bars the mean value for each of the three sets of sensitivity tests. For the sensitivity tests on the parametrization on gas transfer (from 2 to 9), relation with a quadratic (2), hybrid (3 to 5), cubic (6) wind speed dependency are, respectively, in light pink, yellow and orange, and relations that includes explicit bubbles parametrizations (7 to 9) are in dark pink. For the test (14) on calcification processes, the bar indicates the result found for the mean PIC:POC ratio, while the black line indicates the range using the minimum and maximum PIC:POC ratios.

Conclusion:

"Moreover, we displayed that neglecting calcification processes could lead to an over~~under~~estimation by ~~23~~16 to ~~58~~57% of the annual uptake, highlighting the need for the refinement of the model in future studies."

5. Language: I spotted numerous (minor) grammar mistakes, e.g., related to prepositions (see detailed comments below). While this did not impact the readability much, I encourage the authors to carefully check the text again during the revisions.

Response: We warmly thank Reviewer 2 for all the grammar corrections and apologize for these errors. We will carefully check the text again.

Detailed comments:

L. 22: Maybe better: "seasonal and annual budget"?

Response: The sentence will be changed as suggested in the revised manuscript.

L. 26: “reduction of oceanic CO2 uptake”

Response: The sentence will be changed as suggested in the revised manuscript.

L. 27: I suggest rephrasing this sentence by being more specific: Aren’t the physical fluxes (of DIC) always larger than the biological ones? How are both dominant?

Response: The vertical and horizontal fluxes are always both one order of magnitude higher than the net biogeochemical fluxes (respiration minus primary production). However, the net physical flux, i.e. vertical flux plus lateral flux, is of the same order of magnitude as the net biogeochemical flux for each season ; it is higher (in intensity) than the biogeochemical flux in winter and summer (cumulative fluxes: 4.45 versus -1.53 mol C m^{-2} over the winter period, 0.79 versus -0.57 mol C m^{-2} over the summer period), and smaller (in intensity) in fall and spring (cumulative fluxes: 0.49 versus 0.56 mol C m^{-2} over the fall period, -0.80 versus -2.19 mol C m^{-2} over the summer period). At the annual scale, the net physical flux is 3.3 mol C m^{-2} yr^{-1} and represents 88% of the net biogeochemical flux, while the air-sea flux is one order of magnitude smaller and represents 13% of the biogeochemical flux. We will rephrase the sentence, as follows:

“We highlight the ~~dominant~~ major role in the annual dissolved inorganic carbon budget of both ~~biological~~ biogeochemical and physical fluxes that amount to 3.3 mol C m^{-2} yr^{-1} and -3.7 mol C m^{-2} yr^{-1}, respectively, and are one order of magnitude higher than the CO_2 air-sea flux ~~in the annual dissolved inorganic carbon budget~~.”

L. 28: define “upper”

Response: We will specify “upper” as follows:

“The upper layer (from surface to 150 m depth) of the northwestern deep convection region [...]”

L. 29: I suggest replacing “air-sea flux” by “oceanic CO2 uptake”

Response: The sentence will be changed as suggested in the revised manuscript.

L. 37: “comparable role to…” and “processes for carbon”; carbon transfer from where to where? Please specify.

Response: The sentence will be changed for more clarity, as follows:

“Physical mechanisms can quantitatively play a comparable role to that of ~~biological~~ biogeochemical processes on ~~carbon transfer in the ocean~~ CO_2 air-sea flux at regional and global scales …”

L. 42: I suggest rephrasing to “taken up at the ocean surface”

Response: The sentence will be changed as suggested in the revised manuscript.

L. 43: If there is an “on the other hand”, I am immediately looking for “one the one hand”. Maybe better: “at the same time” or “simultaneousy”?

Response: “on the other hand’ will be replaced by “furthermore” in the revised manuscript.

L. 56: moderate phytoplankton bloom

Response: The sentence will be changed as suggested in the revised manuscript.

L. 55-56: Is it typical in the Mediterranean science community to refer to the fall bloom as the “first” bloom? I realize this is a matter of defining the start of the growing season, but from all other regions globally, I am used to describing the bloom phenology starting with the strong first bloom in spring after nutrients were replenished in winter and a secondary typically weaker bloom in the fall.

Response: Some of previous studies which determined the date of the onset in the Mediterranean Sea (Bernardello et al., 2012; Lavigne et al., 2013) were based on the work by Henson et al. (2009) who determined the bloom start in the North Atlantic Sea by adjusting the method of Siegel et al (2002) and considering the 1st September as the beginning of the annual period, to capture the start of the subtropical bloom that occurs in autumn. Using satellite derived-chlorophyll data, Lavigne et al. (2013) found a bloom starting in autumn (late November / early December) in all the Mediterranean bioregions defined by D’Ortenzio and Ribera d’Alcala (2009). Kessouri et al. (2018) calculated the date of the bloom onset in the Western Mediterranean Sea using the same biogeochemical model (without the carbonate system module) as used in this study. They also found a start bloom in autumn for the three considered regions (deep convection zone, shallow convection zone and stratified region). In their results, contrary to the two other regions, in the deep convection region the bloom is interrupted during the deep mixing period and a second bloom start was found when the water column stratified. However, in other studies, the description of the annual chlorophyll cycle is described from January to December and thus the spring bloom is mentioned before the autumnal bloom (Bosc et al., 2004). Our study was performed in the continuity of Kessouri et al. (2018) and thus we preferred keeping the same annual period. In the revised version, we will remove “first” and “secondary” from the sentence.

Henson, S. A., Dunne, J., & Sarmiento, J.: Decadal variability in North Atlantic phytoplankton blooms. Journal of Geophysical Research, 114, C04013. https://doi.org/10.1029/2008JC005139, 2009.

Lavigne, H., D’Ortenzio, F., Migon, C., Claustre, H., Testor, P., Ribera d’Alcala, M., et al.: Enhancing the comprehension of mixed layer depth control on the Mediterranean phytoplankton phenology. Journal of Geophysical Research: Oceans, 118, 3416–3430. https://doi.org/10.1002/jgrc.20251, 2013.

Siegel, D. A., Doney, S. C., & Yoder, J. A.: The North Atlantic spring phytoplankton bloom and Sverdrup’s critical depth hypothesis. Science, 296, 730–733. https://doi.org/10.1126/science.1069174, 2002.

L. 57: “nutrients to the euphotic layer”

Response: The sentence will be changed as suggested in the revised manuscript.

L. 68: Please add a reference to Fig. 1.

Response: The reference to Fig. 1 will be added in the sentence as suggested in the revised manuscript.

L. 71: “which bring DIC-rich water to the surface”

Response: The sentence will be changed as suggested in the revised manuscript.

L. 78: Maybe “complemented” instead of “enriched”?

Response: The sentence will be changed as suggested in the revised manuscript.

L. 79: delete "fixed"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 81: "drives an increase in surface pCO2"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 83: model instead of modelling

Response: The sentence will be changed as suggested in the revised manuscript.

L. 83-84: I am not sure what this approach means. Can you rephrase this part?

Response: In response to one of the main comments and in revising the introduction, this sentence has been simplified as follows:

"D'Ortenzio et al. (2008) and Cossarini et al. (2021), based on a 1D model and a 3D model, respectively, found that the whole deep convection region is a major sink of atmospheric CO_2 *in the open Mediterranean Sea"*

D'Ortenzio et al. (2008) implemented a 1D model in cells of 0.5° x 0.5° horizontal resolution covering the Mediterranean Sea, with no lateral connection between the cells.

L. 86: biological instead of biology

Response: The sentence will be changed as suggested in the revised manuscript.

L. 93: "limited to"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 92-94: This sentence was very confusing to read due to all the “or”. Can you rephrase or split it into two?

Response: In revising the introduction, this sentence has been merged with the following one, as follows:

“In the previous studies, the 3D dynamics of the CO_2 system over an annual cycle has never been specifically explored for the whole northwestern deep convection region and a complete DIC budget is still lacking for this region.”

L. 94-96: To me, this knowledge does not yet become clear enough from what is written up to this point. I suggest revising the introduction to more clearly highlight the knowledge gaps and why these matter.

Response: As we answered at one of the main comments, we will revise the introduction to more clearly highlight the knowledge gaps.

L. 104: “by a positive net community production”

Response: The sentence will be changed as suggested in the revised manuscript.

L. 108: Maybe better: “take advantage of” instead of “benefit from”

Response: The sentence will be changed as suggested in the revised manuscript.

L. 112: Throughout the paper, you sometimes say “biological” and sometimes “biogeochemical”. I suggest to consistently use one because from what I can see (please correct me if I am wrong), you are always referring to the same processes.

Response: The term “biological” will be replaced by “biogeochemical” throughout the paper when referring to the same processes.

L. 120 & L. 125: Have the different models been evaluated in detail over the bigger study regions in any of these studies? It might help to explicitly state that for the interested reader.

Response: The biogeochemical model implemented in the whole Mediterranean and forced by the outputs of the hydrodynamic model NEMO operated by Mercator was assessed by Kessouri (2015) in terms of spatial and temporal surface chlorophyll and vertical distribution of chlorophyll and inorganic nutrient. This will be specified in Section 2.1.1 "The coupled hydrodynamic-biogeochemical-chemical model". The western Mediterranean biogeochemical model was assessed over the western Mediterranean in Kessouri et al. (2018) through comparisons with satellite chlorophyll data.

"The model has been used to study biogeochemical processes in the NW (northwestern) Mediterranean deep convection area (Herrmann et al., 2013; Auger et al., 2014; Ulses et al., 2016; 2021; Kessouri et al., 2017; 2018) and in the whole Mediterranean Sea (Kessouri, 2015)."

Kessouri, F.: Cycles biogéochimiques de la Mer Méditerranée : processus et bilans, Ph.D. thesis, Université Toulouse 3, 2015.

L. 124: How are particle dynamics parametrized in the model? Given that sinking fluxes of biologically-derived particles are an important part of your study, some information on that will be helpful.

Response: To take into account particle dynamics in the model, we consider a constant settling velocity, w_s, for the slow and fast sinking particulate organic matter and for micro-phytoplankton. The values of the settling velocity will be given in Section 2.1.1. The settling of particles is taken into account in the following advection-diffusion equation allowing the calculation of the "physical" rate of change of the concentration C, the concentration of each biogeochemical state variable:

$$\frac{\partial C}{\partial t} + \frac{\partial uC}{\partial x} + \frac{\partial vC}{\partial y} + \frac{\partial (w-w_s)C}{\partial z} = \frac{\partial}{\partial z}\left(K_z \frac{\partial C}{\partial z}\right) + F_C$$

where u, v and w are the three components of the current velocity, K_z is the vertical diffusivity and F_c is the source or sink term from rivers, atmosphere and sediment.

"Particulate organic detritus and microphytoplankton have a constant settling velocity (1 m d^{-1} for slow sinking detritus and microphytoplankton, and 90 m d^{-1} for fast sinking detritus)."

L. 131: Before looking up the cited references, it was unclear to me how the version before can resolve the cycling of carbon without including DIC. I suggest clarifying that only particulate organic carbon was included before.

Response: As suggested by Reviewer 2 we will add a sentence to clarify this point.

"In previous versions of the model, particulate and dissolved organic carbon was considered, but the dynamics of dissolved inorganic carbon was not described."

L. 136: "is the respiration"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 142: "not the case for total alkalinity"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 146: Maybe add "throughout the water column" if that is what it is.

Response: We will add "throughout the water column" in this sentence as suggested in the revised manuscript.

L. 147-149: Personally, I wouldn't call a paper from 2005 "present knowledge". There are several studies that, albeit of course not perfect, have parametrized it. Thus, I suggest rephrasing this part.

Response: This part will be rephrased, as follows:

"The ~~present knowledge on~~ $CaCO_3$ precipitation ~~makes it~~ is difficult to parametrize ~~this term~~ in a model (Aumont et al., 2005). However, we are aware that future refinements will have to take it [...]"

L. 149: "tests on this"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 165: Please add a reference to Fig. 1.

Response: A reference to Fig. 1 will be added in the revised manuscript.

L. 167: I suggest adding "have been described in detail in X and Y and will be summarized here."

Response: The sentence will be changed as suggested in the revised manuscript.

L. 169: It is unclear to me what "hydrodynamic analyses" are. Please clarify and possibly rephrase.

Response: Hydrodynamic analyses represent here the hydrodynamic solutions from the NEMO numerical model computed with the Mercator near real time configuration PSY2V2R4 that embeds assimilation of data in order to constrain and bring realism to the numerical solution. We will replace "analyses" by "fields" in the text.

L. 167-175: I found the description of the steps rather difficult to follow. I think adding a flow chart detailing the different steps could help a lot.

Response: As answered to one of the main comments, to clarify this point we will add a figure with a scheme of the 3 steps in Supplementary material.

L. 179: Given that the model simulates the negative charge and not alkalinity, did you correct the measured alkalinity to correspond to the model tracer? Please clarify.

Response: We apologize for the confusion. Yes, we deduced the initial values of the excess negative charge using measurements of total alkalinity and nutrients concentrations based on Eq. 2. We will add a sentence to clarify this point:

"To deduce the excess negative charge from total alkalinity (Eq. 2), we also used the nutrient concentration data from the Medar/Medatlas database as in Kessouri et al. (2017)."

L. 183: What is a "rigorous mathematical approach"? Please clarify or delete.

Response: We will delete "rigorous mathematical approach" and rephrase the sentence.

"Recently, Davis and Goyet (2021) ~~showed~~ described a ~~rigorous mathematical approach~~ method based upon the property variability, to precisely quantify the uncertainties at any point of an interpolated data field."

L. 189: You only specify what was used for winds here. What about other atmospheric forcing variables (e.g., radiation, humidity, precipitation etc.)? Please be complete.

Response: We will be complete and add all the other forcing variables needed for the gas transfer velocity knowing that the hydrodynamic model uses other atmospheric variables such as air temperature, precipitation, longwave and shortwave radiation:

"To compute the gas transfer velocity, we used the 3-hour wind speed, pressure, and humidity provided by the ECMWF model on a 1/8° grid, in consistency with the hydrodynamic simulation."

L. 191: What I am missing here is a description on the model run time in each step. Also, in L. 179 you mention an initialization in summer 2011, while I think (if I understood correctly), the final model was run from September 2012 onwards. Could you clarify? My confusion on this point convinces me even more that a flow chart detailing the model setup procedure would help.

Response: We apologize for the confusions. The biogeochemical simulation over the whole Mediterranean Sea (step 1b) was performed from 15 June 2011 to 15 November 2013. We initialized the CO_2 system module using interpolated data as it was described L 178-183. The biogeochemical simulation over the western Mediterranean (step 2) was performed over the period from 15 August 2012 to 30 September 2013, and was initialized using the model outputs of the whole Mediterranean Sea simulation. To avoid confusions, as indicated in the response of one of the main comments, we will add the model run time of each step and will move the description of the initialization of step 1b before describing step 2.

L. 193: I find "DIC flows" and "inventory variations" rather confusing. Maybe "DIC fluxes" and "inventory tendencies"? Please check throughout the text.

Response: We will rephrase the sentence and change "flows" by "fluxes" throughout the text. We could replace "inventory" by "stock" or "content" if it is clearer.

"We computed DIC ~~flows~~ fluxes and the resulting content variation ~~in the DIC inventory~~ for the whole deep convection area."

L. 194: "for at least 1 day"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 201: Given the title of this section, I wonder if Eq. 1 is better to be placed here. Additionally, I think at least the general budget equation (Eq. S1) should be moved to the main text.

Response: We would prefer to keep Eq. 1 in Section 2.1.1, since it gives the biogeochemical rate of change of the state variable DIC at the model grid points. As suggested, we will replace the text describing the budget *"The biological term of the budget [...] upper layer is given in Supplementary Material (Text S1)"* in section "Study area and computation of DIC balance" by Text S1.

L. 203: What do you mean by "internal variation"? Please clarify.

Response: "internal variation" meant variation of the content of DIC during a considered period. It is given in Eq. S1 of the submitted version. This term will be removed here. We will also replace it throughout the text.

L. 215: Please add a reference to the respective Equation.

Response: We will add a reference to the Equation in the revised manuscript.

L. 216: "as 0.5"

Response: This sentence will be modified to take into account a comment of Reviewer 1.

"Miquel et al. (2011) estimated the PIC:POC ratio varying between 0.31 and 0.78, with a mean value of ~~to~~ 0.5 at 200 m depth based on sediment trap measurements at the EMSO-DYFAMED site."

L. 220: Please be precise: NCP does not appear as such in Eq. 1.

Response: We agree, the sentence was confusing, We will move "in Eq. 1" as follows:

" [...] we added in Eq. 1 a consumption term representing 35% of NCP ~~in Eq. 1~~. "

L. 221: Please state here what the parametrization by Lajaunie-Salla et al. (2021) is. Ideally, the reader should not have to look up other papers to understand what you're doing.

Response: In Lajaunie-Salla et al. (2021), carbonate precipitation, named *Precip*, is given by the following equation:

$$Precip = k_{precip} \frac{(\Omega_c - 1)}{0.4 + (\Omega_c - 1)} \sum_{i=1}^{3} (GPP_i - RespPhy_i)$$

where k_{precip} is the PIC:POC ratio and Ωc the aragonite saturation, which we set at 3.5 based on Schneider et al. (2007).

We will add the equation in the text:

"In a second sub-test, we added a CaCO3 production term based on the parametrization used in the Gulf of Lion's shelf modeling study by Lajaunie-Salla et al. (2021) (their Table A4, $Precip = k_{precip} \frac{(\Omega_c - 1)}{0.4 + (\Omega_c - 1)} \sum_{i=1}^{3} (GPP_i - RespPhy_i)$, where k_{precip} is the PIC:POC ratio and Ωc the aragonite saturation, set at 3.5 based on Schneider et al. (2007))."

L. 225: sea surface

Response: The sentence will be changed as suggested in the revised manuscript.

L. 284: I suggest adding "reflecting a" in front of "period"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 298: Does the southern zone include everything south of 41°N or is there a southern limit?

Response: The southern zone includes all stations south of the convection zone. There is no southern limit.

L. 299: I assume the depth profiles have been subsampled to only include the cruise locations shown in Fig. 3. Please clarify.

Response: The modeled mean profiles shown in Fig. 5 correspond to the average of the modeled profiles extracted at the same location and date as the measurement stations. This will specify in the text:

"Comparisons were performed by extracting model outputs at the same date and location as measurements."

L. 324: Do you mean "alternating" instead of "alternative"?

Response: Yes, the sentence will be changed as suggested.

L. 325: Where can the direction of the wind be seen? If this is previous knowledge for the region of interest and you therefore decided not to show this explicitly, please make sure it is introduced in the introduction for clarity.

Response: We will specify the wind direction in the introduction, on Figure 1 and/or in Fig. S1 (of the submitted version of the manuscript) by adding two panels with maps of wind speed and direction.

L. 337: Unless I misread something, I think the minus sign should be omitted (the cumulative flux is positive according to Fig. 7).

Response: In fall, the cumulative air-sea flux is negative (Fig. 7b of the submitted version), we are sorry if it was not clear on Figure 7b. As recommended in one of the main comments, we will add a figure with schemes of the seasonal budget for which the direction of the flux will be clearer.

L. 344: Do you mean "DIC concentration in the ML" or "the DIC flux into the ML"? Please clarify.

Response: We meant a decrease in " DIC concentration" visible at the end of October and end of November in Figure 10b. We will slightly modify the sentence, as follows:

"This led notably temporally to ~~a temporal decrease~~ low ~~in~~ DIC concentration into the mixed layer end of October and end of November (Fig. 10b)."

L. 441: “episodes of heat gain”

Response: The sentence will be changed as suggested in the revised manuscript.

L. 466: To me, it is odd to call this flux biological production, when this is in fact remineralization/respiration. I understand why you do it and it is technically correct, but I still suggest rephrasing to avoid confusion.

Response: We agree that “production” can be confusing. We will replace this term by ‘gain” here and a more appropriate term throughout the text and in figures:

“Within the sea, biogeochemical processes induced an annual consumption of 3.7 mol C m^{-2} yr^{-1} *of DIC in the upper layer and a ~~production~~ gain of 2.3 mol C* m^{-2} yr^{-1} *in the deeper layers.”*

L. 469: For consistency with how you described the biological component, it would be easier to read if you also reflected the sign convention in your wording here.

Response: We will modify the sentence to reflect the sign convention as follows:

“Our estimate of net physical fluxes (lateral plus vertical) is an input of 3.3 mol C m^{-2} yr^{-1} *in the upper layer and an export of ~~-~~11.0 mol C* m^{-2} yr^{-1} *in the deeper layer.”*

L. 474: I suggest deleting “an amount”.

Response: The sentence will be changed as suggested in the revised manuscript.

L. 486: Please see my comment on the abstract regarding “both dominate”. I suggest to also rephrase here.

Response: We will also rephrase in the introduction of the discussion section, by merging the two last sentences:

“Our results show that ~~both biological~~biogeochemical and physical processes, ~~dominate the CO2 budget in the upper layer (0-150 m) of the convection zone for the study period., T,~~ through their impacts on DIC concentration, ~~biological and physical flows~~ have both a major role in the intensity and sign of the air-sea exchanges in the deep convection area.”

L. 489: Here and throughout the discussion section: Can you find more descriptive/informative section titles? It is incredibly useful to the reader if the title of each section already conveys information, i.e., ideally the main take-away message.

Response: We will modify titles of the discussion section, as follows:

- ***"5.1 The pCO2" to "5.1 Assessment of the seasonal cycle of the pCO2"***
- ***"5.2 The air-sea CO_2 flux" to "5.2 Estimate of the annual air-sea CO_2 flux and its uncertainties" and "5.3 Comparisons on air-sea CO_2 flux in different Mediterranean regions"***
- ***"5.3 Physical flows in the deep convection area" to "5.4 The major influence of physical transport in the deep convection area"***
- ***"5.4 Net community production and air-sea fluxes" to "5.5 Net community production and air-sea fluxes relationships"***

L. 490-502: As far as I can see, these are results. I am not convinced this part is necessary.

Response: We will remove most of this part. Some elements will be kept to make easier the comparisons with previous studies.

L. 508-509: This sentence is unclear to me. Can you rephrase?

Response: We will rephrase this sentence as follows:

"The high frequency measurements at the CARIOCA buoy described by Hood and Merlivat (2001) and Merlivat et al. (2018) indicated ~~that~~ an interannual variability of 4-5 weeks in the date ~~of the change of sign of~~ at which the pCO_2 difference changes sign, ~~shows interannual variability and is within a period lasting for more than a month~~ depending on air-sea heat flux variations and the timing of the bloom onset."

L. 528-530: Here and throughout the text: Try to avoid 1-2 sentence paragraphs.

Response: We will avoid this as much as possible throughout the text.

L. 552: Please see my major comment on these sensitivity experiments.

Response: As already answered to one of the main comments, we have corrected this error in the equation of the rate of change of alkalinity (excess negative charge denoted $\Sigma[-]$) and have again performed the sensitivity tests. In the new results, the air-sea flux could be reduced by 16% to 57% if carbonate production is taken into account. We will modify the text and Figure 14 in the discussion section on the sensitivity tests, in Sect. 2.1.4 "Sensitivity tests" and in the conclusion.

Discussion section:

They show that not taken into account calcification processes could lead to an ~~underestimation~~ overestimation of the annual air-sea CO_2 uptake by ~~23~~16 to ~~58~~57% with estimates ~~of 0.72~~0.29 mol C m^{-2} yr^{-1}, based on the mean PIC:POC ratio given by Miquel et al. (2011) (varying between 0.20 and 0.36 mol C m^{-2} yr^{-1} based on the maximum and minimum PIC:OC ratios, respectively), and ~~0.58~~0.40 mol C m^{-2} yr^{-1}, based on the parametrization used in Lajaunie-Salla et al. (2021)."

L. 566: Is there a "yr-1" missing? Additionally, it would help to provide the range based on your model here again to compare to the cited paper more easily.

Response: Yes, we will correct the unit by adding a "yr-1" and add in the following sentence the range of the model estimates to make the comparison more easy:

"The larger homogeneity in our estimates (varying between -0.1 and 1.2 mol C m^{-2} yr^{-1} inside the deep convection area) could be partly ascribed to the horizontal diffusion and advection that were accounted for in our model."

L. 576: It might be more appropriate to say "physical transport".

Response: The sentence will be changed as suggested in the revised manuscript.

L. 577: "the vertical DIC distribution"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 581: "greater magnitude" – Please specify the sign.

Response: We will specify the sign:

"They both show a similar seasonal cycle with greater magnitude (positive for the vertical transport and negative for the lateral transport) in fall, the preconditioning phase [...]"

L. 582: "sea heat loss" Do you mean "ocean heat loss"? Please clarify.

Response: We will replace "sea heat loss" by "sea surface heat loss".

L. 589: Please rephrase "DIC exchange flows".

Response: We will replace "DIC exchange flows" by "DIC fluxes at the limits of the deep convection area".

L. 595: "as illustrated in"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 608: "slowed down" instead of "braked"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 617: "convection" instead of "convention"?

Response: We will correct this error in the revised manuscript.

L. 633: "from" instead of "into"?

Response: The DIC budget shows a lateral DIC transport from the surrounding region into the deep convection region in the deep layer (Figure 12). We will change the sentence as follows:

"More specifically, we found that the lateral exchanges with the surrounding region were characterized by a net lateral input of carbon into the deep layers of the deep convection region, [...]"

L. 634: "a lateral outflow"

Response: The sentence will be changed in the revised manuscript.

L. 640-646: It would be a lot easier to compare to the findings of your studies, if you reported these numbers as flux densities instead of as integrated fluxes (or to here report your findings in the same integrated unit).

Response: Our estimate was reported L 645 in the same unit: 0.4 Tg C yr^{-1}. We will slightly change the sentence as follows:

"Thus the NW Mediterranean deep convection area, which represents 2.5% of the Mediterranean Sea surface, and which, we estimate here absorbed at the surface 0.4 Tg C yr $^{-1}$, could strongly contribute to the uptake of atmospheric CO_2 in the open Mediterranean Sea."

L. 648: "into" instead of "in"

Response: The sentence will be changed as suggested in the revised manuscript.

L. 666: I suggest adding "…and rising atmospheric CO2 levels".

Response: We will add this in the sentence as suggested in the revised manuscript.

L. 680: budgets

Response: The sentence will be changed as suggested in the revised manuscript.

L. 691: What exactly are the first and second part here? Please clarify.

Response: The sentence will be changed as follows:

"The region was marked by a deficit of CO_2 compared to the atmosphere from November to early June ~~the second part of fall to the first part of spring~~, which led to a 7-month ingassing of atmospheric CO_2"

L. 701: "subject to"

Response: The sentence will be changed as suggested in the revised manuscript.

Figures:

Fig. 3: Please specify for what depth(s) the model output is shown here.

Response: In the caption, we indicated that the model outputs are "modeled at 3 m depth".

Fig. 4: I suggest adding a legend/title above each column.

Response: We will add "Observations' and "Model" above the first and second column, respectively.

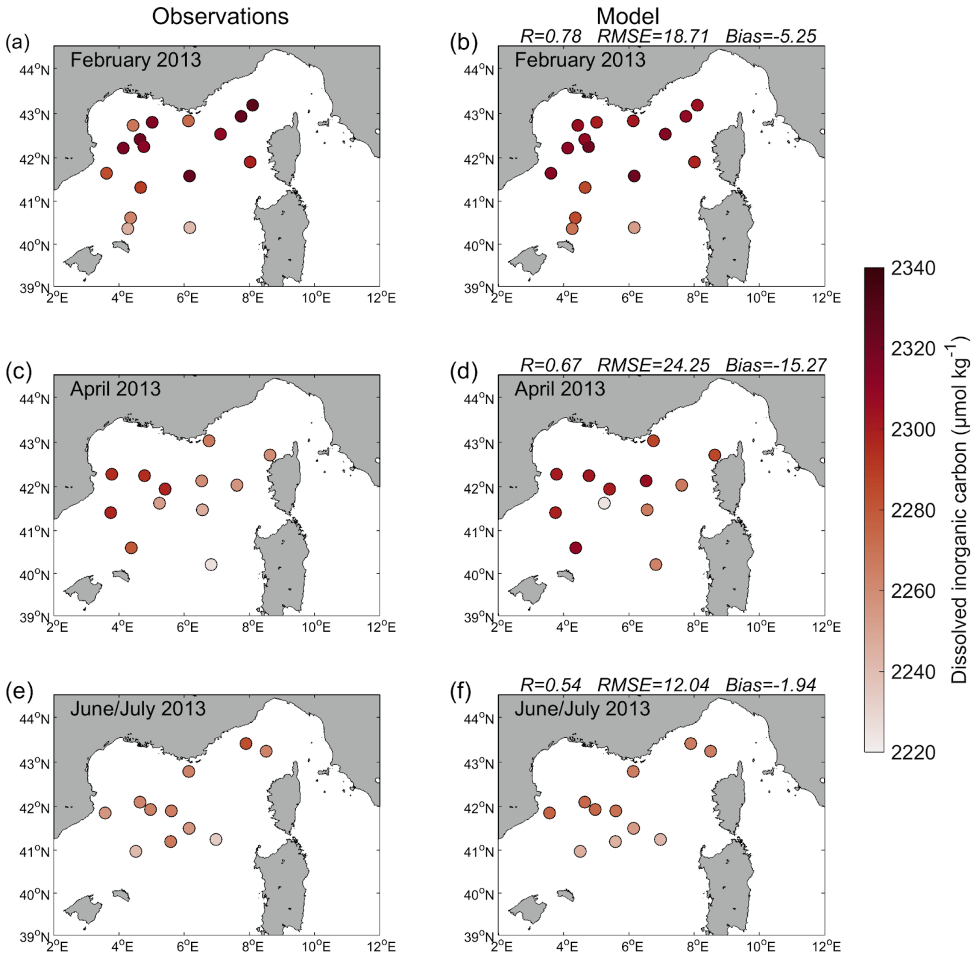


Figure 4: Surface dissolved inorganic carbon (DIC) concentration (μmol kg^{-1}) observed (left) and modeled (right) over the (a,b) DEWEX Leg1 (1-21 February 2013), (c,d) DEWEX Leg2 (5-24 April 2013), and (e,f)

MOOSE-GE (11 June-9 July 2013) cruise periods. The correlation coefficient (R), root mean square error (RMSE), and bias between surface observed and modeled DIC are indicated in (b,d,f).

Fig. 7: I suggest using the same colors for the same components in all panels, not only in a & b, but also in panel c. Additionally, it is unclear to me why you decided to show the seasonal averages only for the upper layer and not for the deeper layer. Please consider adding the extra panel for completeness.

Response: The color for the different components in Fig. 7 was the same color as the same components shown in Fig. 6:

- biogeochemical fluxes in the upper layer in bright green,
- biogeochemical fluxes in the deeper layer in green/brown,
- physical fluxes in the upper layer in light blue,
- physical fluxes in the deeper layer in dark blue.

As recommended in one of the main comments, we will add a figure with seasonal budget schemes showing the budget in the upper and deeper layer, and to avoid repetitions we will remove Fig 7b.

Fig. 14: Please link the caption more clearly to the figure: which bar is which experiment? Only giving the reference requires the reader to be familiar with every single paper, which will not necessarily be the case (it certainly isn't the case for me).

Response: To clarify this figure, we will move the titles of the experiment in the top of the figure. We will also classify and color the bars according to the type of parametrization of the gas transfer velocity instead of the date of paper publication for the first set of experiments, and we will add the type of the parameterization in the caption.

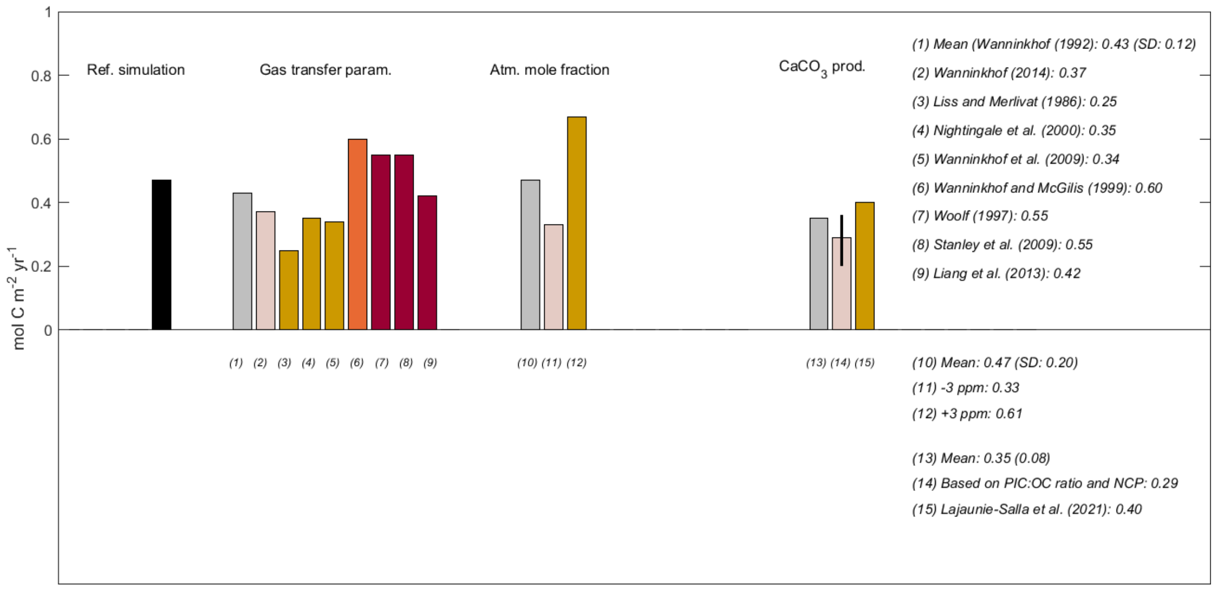


Figure 14: Sensitivity tests to the parameterization of gas transfer velocity, the variability of the mole fraction of CO_2 in the atmosphere, and the calcification processes on the annual CO_2 air-sea flux estimate. The black bar indicates the annual estimate in the reference simulation, grey bars the mean value for each of the three sets of sensitivity tests. For the sensitivity tests on the parametrization on gas transfer (from 2 to 9), relation with a quadratic (2), hybrid (3 to 5), cubic (6) wind speed dependency are respectively in light pink, yellow and orange, and relations that includes explicit bubbles parametrizations (7 to 9) are in dark pink. For the test (14) on calcification processes, the bar indicates the result found for the mean PIC:POC ratio, while the black line indicates the range using the minimum and maximum PIC:POC ratios.

All figures: Please double-check that the sign convention of all fluxes is defined in the respective caption.

Response: We will check this.

Supplementary material: Eq. S2: "DCA" is not defined in the text.

Response: DCA was defined in L. 5 of the Supplementary material. We will move its definition just after the equation (that will be moved in the main text as recommended in a previous comment):

"where (x,y,z) belongs to the upper layer (150 m to the surface) of the DCA (deep convection area)."

L. 29: How are sediment fluxes treated in the model? How large are they compared to the other components? Without any further information, it is difficult to judge for the reader to what extent this assumption impacts the role of vertical fluxes (which are treated as the residual and will therefore include any sedimentary contribution).

Response: The fluxes of dissolved inorganic carbon, nutrients and oxygen at the sea-sediment interface were calculated using a simplified version of the vertically-integrated dynamic sediment model described in Soetaert et al. (2000). The parameters of the model were set following the study of Pastor et al. (2011) in the Gulf of Lion shelf. The same model was used by Many et al. (2011) who showed that the model results were consistent with previous observational and modeling studies on the Gulf of Lion shelf. In this study, we found a POC deposit of 0.1 mol m^{-2} yr^{-1} in the deep convection area. This is in the same order, but smaller than the sediment flux estimated at 0.2 mol C m^{-2} yr^{-1} by Stabholz et al. (2013) near the bottom in the deep convection area. The authors reported an increase of the flux by one to two orders of magnitude during a winter characterized by deep convection. They attributed this increase to resuspension events induced by strong bottom currents. Durrieu de Madron et al. (2023) also pointed out the influence of dense shelf water cascading which can be responsible for supplementary organic carbon deposit flux. In the model, the efflux of DIC resulting from the sediment organic carbon remineralization is calculated during the simulation and taken into account in the budget but is negligible compared to all the other terms. Further comparison analyses will be needed in the future to verify the model in the deep region. Moreover, a coupling with sediment transport model would allow improving the description of the deposition flux of organic carbon and the modifications in the sediment resulting from resuspension events. In the revised manuscript, we will specify how the fluxes are calculated at the sea-sediment interface and indicate that we found a negligible annual DIC flux.

Durrieu de Madron X., D. Aubert, B. Charrière, S. Kunesch, C. Menniti, O. Radakovitch, and J. Sola. 2023. Impact of dense water formation on the transfer of particles and trace metals from the coast to the deep in the northwestern Mediterranean. Water, 15, 2: 301. doi: 10.3390/w15020301.

Stabholz, M., Durrieu de Madron, X., Canals, M., Khripounoff, A., Taupier-Letage, I., Testor, P., Heussner, S., Kerhervé, P., Delsaut, N., Houpert, L., Lastras, G., and Dennielou, B.: Impact of open-ocean convection on particle fluxes and sediment dynamics in the deep margin of the Gulf of Lions, Biogeosciences, 10, 1097–1116, https://doi.org/10.5194/bg-10-1097-2013, 2013.