Interactive comment on "Coastal-ocean uptake of anthropogenic carbon" by Timothée Bourgeois et al.

We thank Prof. Pierre Regnier (referee #1) and Prof. Nicolas Gruber (referee #2) for their comments and suggestions. We also thank Prof. Jack Middleburg for managing the peer-reviewing process of this manuscript. We first present our Author' Response with a point-by-point response to the reviews with relevant changes made in the revised manuscript, with referee comments in grey and author responses in black. Then we present the marked-up manuscript with additions and changes in **bold** and removals in **bold strikethrough**.

#### Point-by-point response to the reviews with relevant changes made in the manuscript

We first comment on 3 key topics raised by the referees, and then provide point-by-point responses for each referee with changes made in the manuscript. The 3 key topics are (1) the need for more information to support our proposed mechanism for lower coastal anthropogenic  $CO_2$  uptake due to limitation by cross-shelf exchange, (2) the need to clarify our estimate of anthropogenic carbon fluxes and on the role of potential biological changes, and (3) the effect of model drift on our results.

#### 1) Cross-shelf exchange

We concur with both referees that more information is needed to support our conclusion that it is inadequate cross-shelf exchange that reduces the coastal ocean's uptake of anthropogenic CO2 (per unit area) relative to that of the global ocean.

Following the suggestion by Referee #1, we plan to include a new figure (Figure 10) with time series of dissolved inorganic anthropogenic carbon ( $DIC_{ant}$ ) storage and anthropogenic  $CO_2$  uptake with cross-shelf export of  $DIC_{ant}$ . This new figure demonstrates clearly that simulated cross-shelf export of  $DIC_{ant}$  is less than the simulated anthropogenic  $CO_2$  uptake for the coastal ocean, implying an accumulation of  $DIC_{ant}$  in the coastal waters column during the simulation.

As suggested by the Referee #2, we offer more detail about the simulated coastal-ocean ocean exchange. For that, we added the simulated water residence time for each MARCATS region to Tables 2 and 3 of the revised manuscript and we introduced a new figure (Figure 8). These new results reveal that simulated residence times for most coastal regions are of the order of a few months or less, except for Hudson Bay, the Baltic Sea and the Persian Gulf. The latter three regions are generally more confined and we would expect longer residence times, although our model simulated residence times are shorter than what has been published for similarly defined coastal regions (see Jickells et al. (1998), Men et al. (2015)). The absence of tides and the spatial resolution used in our study do not permit to reproduce the mesoscale variability of the coastal circulation (e.g. eddies and upwellings). Depending on coastal regions, these processes may increase/decrease residence time. The comparison with these published estimates is also difficult since we average residence time estimates on extended areas.

Page 9 Line 8: We added the section 2.6 in the Methods section to explain the residence time calculation:

"2.6 Residence time

To compute water residence time in each MARCATS region, we divided the volume of each region by the integrated outflow of water from 5-day mean current velocities at coastal boundaries."

Page 15 Line 19: We discussed simulated residence time as follows:

"The accumulation in the coastal ocean is effective over the entire period (1910-2012) as the uptake of anthropogenic carbon by the global coastal ocean is always inferior to its cross-shelf export (Fig. 10). To gain insight into this cross-shelf exchange, we computed the simulated mean water residence times for each MARCATS region (Fig. 8). Residence times for most coastal regions are of the order of a few months or less, except for Hudson Bay, the Baltic Sea and the Persian Gulf. The latter three regions are generally more confined and we expect longer residence times, although our model simulations were never designed to simulate these regions accurately. Generally, our simulated residence times are shorter than what has been published for similarly defined coastal regions although methods differ substantially (Jickells, 1998; Men and Liu, 2014; Delhez et al., 2004). Despite these short residence times, the cross-shelf export of anthropogenic carbon is unable to keep up with the increasing air-sea flux of anthropogenic carbon (Fig. 10). This may be explained by the open-ocean waters that are imported to the coastal ocean being already charged with anthropogenic carbon, thus limiting further uptake in the coastal zone."

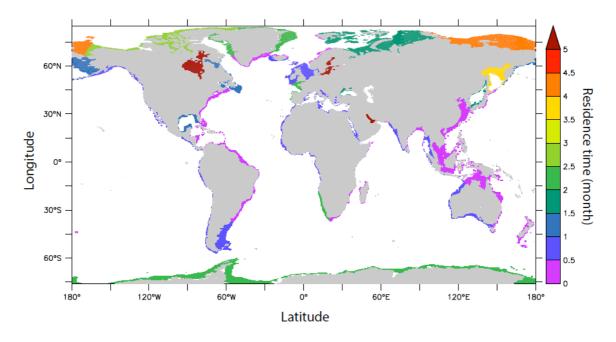


Figure 8. Global distribution of simulated residence time (month) for the global coastal ocean segmented following Laruelle et al. (2013).

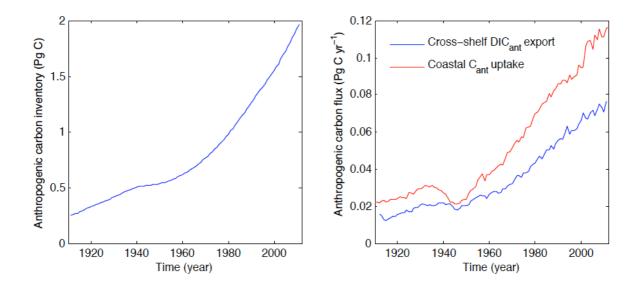


Figure 10. Simulated temporal evolution of (a) coastal-ocean inventory of anthropogenic carbon given in Pg C and (b) anthropogenic CO<sub>2</sub> (C<sub>ant</sub>) uptake by the global coastal ocean and global cross-shelf export of anthropogenic carbon (DIC<sub>ant</sub>) given in Pg C yr<sup>-1</sup>.

### 2) Our estimate of anthropogenic carbon fluxes and the role of potential biological changes

Our method to compute anthropogenic air-sea  $CO_2$  fluxes (FCO<sub>2</sub>) is based on the difference between total FCO<sub>2</sub> from an historical simulation (with increasing atmospheric CO<sub>2</sub>) and natural FCO<sub>2</sub> from a control simulation (i.e., with the same physical forcing but with fixed pre-industrial atmospheric CO<sub>2</sub>). This method is commonly used by the modeling community to estimate anthropogenic carbon fluxes (e.g. <u>http://ocmip5.ipsl.jussieu.fr/OCMIP/</u> or Bopp et al., 2015). By definition, our anthropogenic air-sea CO<sub>2</sub> fluxes only respond to increasing atmospheric CO<sub>2</sub>. They do not include any effect from potential changes in ocean physics or biology, because those changes are identical in the historical and the control simulations. Hence, even if surface temperature or biological fluxes (NEP or NEC) change in response to the forcing, they do not impact anthropogenic carbon uptake per se.

That said, we agree with the reviewers that potential changes in the physics and biology as well as changes in riverine input or in the interactions with the sediment may be of primary importance, would modify the distribution of total carbon and alkalinity, and hence would also modify the potential of the coastal ocean to absorb anthropogenic carbon.

These points are discussed in more detail in the revised manuscript.

Page 2 Line 13: We added "Indeed, in addition to the effect of increasing atmospheric CO2, potential changes in coastal ocean physics (e. g., temperature) and biology (e.g., NEP) as well as changes in riverine input and interactions with the sediment may be of primary importance (Mackenzie et al., 2004; Hu and Cai, 2011). These changes would modify the distribution of carbon and alkalinity, and hence change the potential of the coastal ocean to absorb anthropogenic carbon."

To clarify our method at the end of the Introduction section:

Page 4 Line 4: We added "We focus solely on the geochemical effect of anthropogenic CO2 addition from the atmosphere to the ocean and neglect the role of varying river input and interactions with the sediment, as well as the feedback from a changing climate."

To clarify our method in the Methods section:

Page 6 Line 34: We added "Here, we use the conventional definition of anthropogenic carbon the used previous global-ocean model studies (OCMIP. in ocean by http://ocmip5.ipsl.jussieu.fr/OCMIP/ and e.g., Bopp et al. (2015)), namely that anthropogenic carbon comes only from the direct geochemical effect of increasing atmospheric CO2 and its subsequent invasion into the ocean. By definition, this anthropogenic FCO2 does not include any effect from potential changes in ocean physics or biology. In the model, there are no changes nor variability in riverine delivery of carbon and nutrients, and anthropogenic carbon is not buried in sediments."

Bopp et al. (2015) http://dx.doi.org/10.1002/2015GL065073

#### 3) Model drift and potential implications.

We concur with both reviewers that details about model drift are important. In the original manuscript we stated the following: "At lower resolution (ORCA2), after a spin up of 3000 years, there is 0.26 Pg C yr-1 greater globally integrated sea-to-air flux, relative to results after only a 50-year spin up. Nearly all of that enhanced sea-to-air CO2 flux due to the longer spin up comes from the Southern Ocean." Unfortunately, such spin-up length is currently out of reach regarding to computation costs of our ORCA05-PISCES configuration. We emphasize though that our anthropogenic FCO<sub>2</sub> estimates are expected to be influenced very little by model drift because of the way anthropogenic carbon is defined ( $C_{ant} = C_{total} - C_{natural}$ ), i.e., drift affects both C<sub>total</sub> and C<sub>natural</sub> in the same way.

Page 7 Line 14: We added "Anthropogenic FCO2 estimates are expected to be influenced very little by model drift because of the way anthropogenic carbon is defined, i.e., drift affects both natural carbon and total carbon in the same way."

#### POINT-BY-POINT REPLY

#### • <u>Referee #1 comments</u>

The manuscript by Bourgeois et al. is the very first attempt to quantify the air-sea  $CO_2$  flux for the global coastal ocean using a highly-resolved 3D model. The authors compare in a convincing way their model results with observational data and discuss in detail the obtained spatial variability in the air-water  $CO_2$  exchange. The approach is well described and model results are solid; I am thus very supportive of this research. In addition, the authors have attempted a quantification of the anthropogenic perturbation on the air-sea  $CO_2$  flux, with the key finding that the magnitude of the perturbation could be significantly smaller than previously taught. This is obviously an important result that further strengthens the value of this contribution. However, the latter aspect has several shortcomings that I believe need to be addressed fully (see in particular major comments 2) and 3) before publication.

#### Thanks for this positive general feedback.

#### Major comments

1) Uncertainties are only reported once for the anthropogenic  $CO_2$  flux (0.1 +- 0.01). You need to explain how this uncertainty has been estimated. It is also much lower than the uncertainty on the total simulated flux (0.27 +- 0.07), which is quite surprising. More generally, uncertainties and their quantification method should be reported for all fluxes and consistently throughout the text.

The uncertainty on our estimate of the coastal-ocean uptake of anthropogenic carbon is defined as its interannual standard deviation over 1993-2012. This will be clarified to the revised manuscript; uncertainties are also given for all fluxes (see updated Tables 2 and 3 at the end of our response).

Since our uncertainty values are based on the interannual variability over 1993-2012, the uncertainty on the simulated total  $FCO_2$  is much higher than that of the anthropogenic flux. That is, the strong variability of total  $CO_2$  flux is due almost entirely to variability in its natural component so that the variability in their difference (anthropogenic flux) is small.

Another way to estimate uncertainties for such fluxes would be to use multiple models or sensitivity tests with one model where key parameters were varied. These exercises are currently out of reach and left for future work.

To clarify the definition used for uncertainties:

Page 8 Line 31: We formulate the sentence as is. "In addition, the model's uncertainty, computed as the interannual variability over 1993–2012, is compared to uncertainties in the observational estimates, computed as the standard deviation between flux parameterizations from Wanninkhof (1992) as modified by Takahashi et al. (2009), Ho et al. (2006) and Wanninkhof (1992)."

2) Section 4.1.2 on anthropogenic fluxes provides a suitable comparison with previous estimates. However, the last paragraph is misleading as one of the key reason why the size of the perturbation could be larger in Mackenzie and co-workers is the stimulation of the biological pump by enhanced land-derived nutrient inputs. These aspects should be included in the discussion, but also much earlier in the text (introduction and, eventually, title). That is, the authors should clearly state right from the start that they only consider atmospheric  $CO_2$  as their sole anthropogenic driver. As a result, I believe that only the physical dissolution pump is impacted, i.e., the model should simulate constant net ecosystem productivity (NEP) and (I suspect) constant net ecosystem calcification (NEC) during the entire historical period. The values for NEP and NEC should be reported and discussed (a subject of intense debate within the coastal C community) as this could be (another) plausible reason for the discrepancy with earlier estimates. Finally, nothing is said about temperature effects on the uptake of  $CO_2$ . This aspect should also be included in the description/discussion.

We agree with the reviewer that the way we define the anthropogenic flux hampers any strict comparison with some previous estimates in which other terms are taken into account. This

## will be clarified in the revised manuscript, and we refer the reviewer here only to our general reply above for a first response to this comment.

3) Section 4.2 provides an explanation for the smaller relative magnitude of the global coastal anthropogenic CO<sub>2</sub> uptake compared to the global ocean. As it is, Figs. 2, 5 and 8 do not satisfactorily substantiate the proposed mechanism. What are missing are plots of temporal evolution of (organic and inorganic) carbon accumulation (also % relative increase) and cross-shelf export for the entire simulation period. The Revelle factors should also be reported. Based on the proposed mechanism, I would suspect to see a progressive decrease of the ratio of anthropogenic carbon uptake of the coastal ocean to the global carbon uptake due to the accumulation of anthropogenic  $CO_2$  in the coastal water column through time and this does not seem to be the case (Fig.2). I also would suspect to see a progressive increase in the Revelle factor (faster for the coastal ocean than open ocean) through time. In addition, it would be interesting to briefly discuss why the uptake fluxes per unit surface area for the shelf seem to be larger than for the open ocean under pre-industrial conditions (Fig. 5). Furthermore, the authors should report the calculated horizontal cross-shelf transport of water as this is a crucial number to sustain their conclusion (a first sensitivity analysis could have been useful in this context). Finally, I believe that comment 2 above (focus on the physical dissolution pump only) is also relevant in the context of section 4.2

We concur with the reviewer that more information is indeed needed to substantiate our proposed mechanism; we refer to the general reply (point 1, above) in which we indicate the additional figures and discussions that we introduced in the revised manuscript.

We have also calculated the evolution of the Revelle factor for the coastal and open ocean over the historical period. While the coastal Revelle factor is 16% more than in the open ocean in 1870, that ratio evolves to being only 17% more for the last simulated decades (around 2000). Thus different rates of change of Revelle factor between the coastal and open ocean cannot solely explain the amplified anthropogenic carbon accumulation in the water column of the coastal ocean.

Finally, concerning the horizontal cross-shelf transport of water, we do not fully understand the suggestion of the referee. We have chosen to compute water residence times for each MARCATS. We refer the reviewers to our general reply for this comment.

#### Other comments

Abstract and conclusion: the authors should also summarize the main results on the total fluxes as this is the first time that a model-data comparison is performed with a physically-resolved model at the global scale.

As proposed, the main results on the model-data comparison of total fluxes will be added to the abstract and conclusion as follows "Evaluation of the simulated air-sea fluxes of total CO2 for 45 coastal regions gave a correlation coefficient R of 0.8 when compared to observation-based estimates."

Abstract page 1, line 8: a high resolution is required not only to resolve the bathymetry, but also the complex coastal currents (which in my opinion are not all induced by the bathymetry)

## "To begin to better resolve coastal bathymetry" is replaced by "to better resolve coastal bathymetry and complex coastal currents" in the revised manuscript.

Page 2 line 4: I suspect that the word export refers to 'export production'. I would clarify because in the context of this paper, it could also refer to 'cross-shelf export'

# As the intended meaning was "cross-shelf export", "carbon export" is replaced by "primary productivity, export production and carbon burial" to the revised manuscript to clarify this sentence.

Page 2 line 5: the carbon export and burial fluxes are highly uncertain – see, e.g. Krumins et al. 2013 (BG) for a review. The same also holds for the productivity (even the sign of the NEP is uncertain – see Bauer et al., 2013. It is thus not correct to state that the air-sea  $CO_2$  flux is the most uncertain of the C fluxes for the coastal ocean.

We presume that the referee refers to page 2, line 9 rather than line 5. We agree that the "less is known" formulation for coastal-ocean air-sea  $CO_2$  exchange is not suitable here. In the revised manuscript, the last sentence of this paragraph is replaced by "All these estimates suffer from high uncertainties as do those for coastal-ocean air-sea  $CO_2$  exchange (Laruelle et al., 2014), particularly its anthropogenic component."

Page 2 line 15-16: I agree about the  $CO_2$  switch of the coastal ocean from source to sink, but do not agree fully with the proposed attribution. Mackenzie and co-workers highlight the change in NEP (from enhanced land nutrient inputs) as one of their key driver to explain the shift (see also Regnier et al., 2013 & Bauer et al., 2013 – for reviews). Please clarify (see also major comments).

Referee #1 does well to emphasize the need to introduce the proposed role of the changing NEP and riverine nutrient inputs in the  $CO_2$  source-to-sink shift of the coastal ocean from Mackenzie et al. (2004). This proposed mechanism will be discussed in the revised manuscript as will be that from Bauer et al. (2013) to explain the  $CO_2$  source-to-sink shift due only to the increased physical uptake of atmospheric  $CO_2$  (with constant NEP).

To better introduce the next modifications:

Page 2 Line 25: We added "Therefore, estimates of anthropogenic carbon uptake by the global coastal ocean rely mainly on modelling, extrapolations from the open-ocean and/or closing- or/ balanced- budget approaches. An early modelling approach was proposed by Andersson and Mackenzie (2004) and Mackenzie et al. (2004)."

To take into account referee #1 comment:

Page Line 30: We added "They estimated that the preindustrial coastal ocean was a source of CO2 to the atmosphere and had recently or will switched to a CO2 sink. This source-to-sink switch is mainly caused by a shift in net ecosystem production (NEP) due to increased anthropogenic nutrient inputs (Andersson and Mackenzie, 2004; Mackenzie et al., 2004). Another proposed mechanism is simply linked to the anthropogenic increase in atmospheric CO2, considering constant NEP (Bauer et al., 2013)."

Page 3, line 20: I recommend making reference to the few published long time series of  $CO_2$  observations (> 1 decade) for the coastal ocean. I agree nevertheless that these time series alone are sparse and short. Thus, an observation-based global extrapolation of the anthropogenic component is highly uncertain.

Two references dealing with long time series of  $CO_2$  observations in the coastal ocean were added in the revised manuscript (Astor et al., 2013 with 1996-2008  $CO_2$  data at CARIACO station on Venezuelan coasts and Ishii et al., 2011 with the 1994-2008  $CO_2$  data along 137°E on Japanese coasts) to support the lack of data-based estimates. To our knowledge, these are the only available time series of  $CO_2$  data of more than a decade.

Page 2, Line 20: We added "Estimating air-sea fluxes of anthropogenic CO2 in the coastal ocean would require multidecadal time-series of coastal CO2 observations in order to extract an anthropogenic signal from the strong coastal natural variability. Such time-series are still rare and probably not long enough. To our knowledge, the only available equivalent time-series are the Ishii et al. (2011) 1994-2008 time series along 137°E on Japanese coasts and the Astor et al. (2013) 1996-2008 time-series at the CARIACO station on Venezuelan coasts."

Astor et al. (2013) http://dx.doi.org/10.1016/j.dsr2.2013.01.002 Ishii et al. (2011) http://dx.doi.org/10.1029/2010JC006831

Page 4 line 24: calcite particles are included. This is not a satisfactory description. Please state clearly if your model accounts for calcification as this process has a potentially important impact on the air-sea  $CO_2$  exchange (see also major comments).

We have chosen not to provide great detail on the model's calcification-related processes, which are described extensively elsewhere (Aumont and Bopp, 2006). The simulated NEC remains constant at a first order and does not respond to increasing anthropogenic CO<sub>2</sub>. Moreover, calcification is identical in our historical and control simulations; hence, it does not affect the simulated anthropogenic carbon perturbation.

Page 4 line 29: do you mean atmospheric deposition?

#### Yes, "budgets" is replaced by "deposition" in the revised manuscript.

Page 6 line 4: Assuming that land derived DOC is entirely labile is a strong assumption. The flux (0.15 PgC yr-1 from the top of my head) is also significant. Thus, the extent to which your results depend on this assumption has to be discussed.

Referee #1 is absolutely right that the representation of riverine DOC input in the model is extremely simple. Dissolved organic matter is indeed assumed to remineralize instantaneously at the river mouths, thus contributing to the DIN, DIP, DIC pools. Yet complexifying the approach will not affect our estimate of anthropogenic carbon uptake since the land-derived carbon delivery and its lability remains constant throughout simulation. We agree with the referee on the need to clarify this point in the revised manuscript.

Page 6 lines 11-19: The implication of a model outside of ' equilibrium ' has to be addressed. For instance, when you refer to a global ocean anthropogenic uptake of 2.3 PgC/yr-1, this number is obtained with a natural flux of -0,33 PgC yr-1 for the natural flux. Correct?

#### This is correct. We refer the reviewers to the general reply for this comment.

Page 7, section 2.4 evaluation dataset: To leave no ambiguity, did you compare your model results with LA14 using the Wanninkhoff 1992 formulation or the updated formulation?

We used Laruelle et al. (2014) estimates computed using the formulation of Wanninkhof (1992) as modified by Takahashi et al. (2009) to compare with simulated  $CO_2$  fluxes computed using the initial Wanninkhof et al. (1992) formulation. Although the gas exchange formulation is critical for air-sea flux estimates derived from observations, it has little impact on model-derived fluxes of anthropogenic carbon (Sarmiento et al., 1992).

#### Sarmiento et al. (1992) http://dx.doi.org/10.1029/91JC02849

Page 9, lines 12-14: I would say 'weak carbon sources' and 'strong carbon sinks'

#### We made both of these changes in the revised manuscript.

Page 9 lines 15-25: The phrasing is misleading ('our model results tend to underestimate total carbon flux, with 76% of the simulated specific fluxes lower than the data-based estimates'), as the absolute fluxes are actually larger in the model (i.e. larger negative sinks). 'Likewise' is also not appropriate because the Arctic region is in fact the only latitudinal band where the model results predict a smaller sink than the observations. More broadly, I find that the results are quite comparable for the southern hemisphere and the low latitude regions, but that discrepancies are significantly larger in the Northern hemisphere with a stronger sink modeled for the 30-60° N and a weaker sink modeled for the > 60°N latitudinal band (see also Fig 3 of LA14). Also, the fact that the areal-integrated fluxes show a weaker obs-model correlation than the fluxes per unit surface area requires discussion.

The sentence "our model results tend to underestimate total carbon flux, with 76% of the simulated specific fluxes lower than the data-based estimates" is replaced by "our model results tend to simulate larger sinks and weaker sources than observed (i.e. 76% of the specific simulated fluxes of total carbon have lower relative values than the data-based estimates)".

"Likewise" is replaced by "Otherwise".

Using the latitudinal distribution of natural fluxes, we can explain why carbon uptake is indeed larger in the coastal ocean. This is because upwelling zones (natural  $CO_2$  sources) are extremely restricted (i.e. narrow continental shelf) contrary to the  $CO_2$  sink regions with commonly large continental shelves.

Concerning the weak obs-model correlation for area-integrated fluxes, remapping errors between MARCATS surfaces in Laruelle et al. (2014) and ours are particularly important for low area regions and reach 50% as maximums. Thus, we estimate that the model-data comparison using area-integrated  $FCO_2$  is not adequate with such large remapping errors. We

removed this part of the model-data comparison and the  $2^{nd}$  panel (b) of the Figure 6 in the revised manuscript.

Page 10 Line 18: We removed "but only 0.5 for area-integrated fluxes."

Page 10 line 14: what do you mean by 'top two regions'?

## We replaced "top two regions" by "the most efficient regions in anthropogenic carbon uptake".

Page 10 line 25 and further: It is important to state that (to my knowledge) only LA14 accounts for the sea-ice cover in the global estimates - this is an important effect on the quantification.

We emphasized that the Laruelle et al. (2014) is the first and only study to provide coastal observational-based  $FCO_2$  estimates at global scale taking into account the effect of sea ice. Remarks are added both to section 2.4 (Evaluation dataset) and to section 4.1.1 (Total fluxes) as follows.

Page 8 Line 24: we added "LA14 is the first and only study to provide coastal-ocean observation-based FCO2 estimates at global scale taking into"

## Page 12 Line 2: We added "LA14 is the first observation-based study to take into account this sea-ice effect for coastal-ocean FCO2 estimates at global scale"

Page 11 line 5-10: I agree that the exclusion of the proximal zone in the model assessment should have an impact on the sign of the flux under pre-industrial conditions. But what about the effect of the initialisation (the value of the sink is not reported for the coastal ocean in 1850)? Stated differently, is the global coastal ocean in equilibrium at the onset of the simulations? Regarding the proximal zone, bays, estuaries, deltas, lagoons are indeed sources of  $CO_2$  (see Laruelle et al., 2013 for the latest synthesis), banks should be too (a reference would be useful), but I am not sure about what is meant by ' marine wetlands '. If this refers to marshes and mangroves, they are then believe to be sinks for atm  $CO_2$  (see Cai, 2011, Regnier et al., 2013, Bauer et al., 2013). Thus, clarification is required here.

Certainly, the coastal ocean is not at equilibrium when the simulation is initialized in 1870. This is an important point addressed in our previous responses.

"Marine wetlands" is replaced by "salt marshes and mangroves" in the revised manuscript to clarify the sentence and "banks" will be added to the revised manuscript. Those regions are included in the proximal zone that is generally known as a carbon source although some parts of it may be sinks.

Page 11, section 4.1.2 first paragraph: I believe that regional scale studies have attempted an estimation of the anthropogenic  $CO_2$  uptake in EBUS. If true, they should be included in the discussion.

We have searched but fail into find regional-scale studies that provide estimates of anthropogenic  $CO_2$  uptake in EBUS regions. Estimates of anthropogenic carbon content in the water column have been published for instance for the California current region by Feely

## et al. (2008) but we do not found any regional estimates of anthropogenic $CO_2$ uptake from the atmosphere.

Page 11 line 21-22: This sentence has to be rephrased as it implies that one modeling approach performs better than another. Please tone down.

We do not understand Referee #1's remark to tone down the sentence on line 21-22. Our affirmation that Wanninkhof et al. (2013) exploit coarse-resolution model and data is valid. According to our model, the extrapolation technique used by Wanninkhof et al. (2013) overestimates the anthropogenic carbon uptake of the coastal ocean. We left the sentence as is.

Page 12, section 4.2: the first two paragraphs on total fluxes should be merged with section 4.1.1 – Regarding the Amazon, what is the potential impact of assuming that all the DOC (a large flux) is transformed into DIC in this region? More generally, do you assume that this instantaneous transformation has no impact on alkalinity?

We chose to design the section 4.2 to highlight contrasts between the coastal and the open ocean whereas 4.1.1 is specifically focused on coastal total FCO2. As the first two paragraphs of the section 4.2 deal with coastal vs. open ocean comparison, we suggest letting the paragraphs as is. To clarify the aim of section 4.1, we renamed it as "Comparison with previous coastal estimates".

Our assumption of the instantaneous remineralisation of all land-derived DOC into DIC impacts natural  $FCO_2$  (and total  $FCO_2$ ) but has no effect on simulated anthropogenic  $FCO_2$ . One of the potential of this assumption would be a total  $CO_2$  sink reduction, shown for the Amazon plume for instance. In our model, river alkalinity input is equal to the initial riverine DIC input. But when riverine DOC is remineralized to DIC, that does not affect simulated ocean alkalinity. This point is clarified in the model description.

Page 12 Line 15: We added "The model representation of riverine DOC input and its instantaneous remineralization has potential implications for our estimates of total FCO2. In the Amazon plume for instance, we underestimate CO2 absorption because of this instantaneous addition of DIC without input of alkalinity. However this assumption has no direct implication on our anthropogenic FCO2 estimates."

Page 12, section 4.2: the latitudinal trends in anthropogenic  $CO_2$  fluxes are also very similar for the coastal and open ocean (Figure 5). This aspect needs to be discussed.

The latitudinal distributions of anthropogenic  $CO_2$  fluxes are indeed similar between the coastal and open ocean. In particular, we note that this similarity is prominent in the Southern Ocean: Antarctic shelves and adjacent open ocean waters are very much alike. Following the Laruelle et al. (2014) definition of the Antarctic Shelves, the bathymetry of this coastal region is deeper than the other MARCATS regions. Its mean bathymetry is around 500 m against 160 m for the global coastal ocean. This mitigates the contrast between coastal and open ocean processes in the Southern Ocean. This explanation is added to the revised manuscript.

Page 13 Line 33: We added "Yet the pattern for anthropogenic CO2 flux differs greatly from that of natural CO2, having its strongest uptake in the Southern Ocean in both the open and coastal oceans, i.e., where zonally averaged specific uptake reaches up to  $1.5 \text{ molCm}^{-2} \text{ yr}^{-1}$ .

The bathymetry of MARCATS regions around the Antarctic continent is much deeper than in the other coastal regions (500 m vs. 160 m for the global coastal ocean); this probably reduces the contrast between the coastal and open ocean in the Southern Ocean and explains the similarities of anthropogenic carbon uptake rates there."

Page 13, lines 12-19: The computation of Revelle factor values is interesting, but it is important to stress that (to my knowledge), a higher value for the global coastal ocean compared to the global ocean remains highly speculative as this has not been demonstrated from observational data. Also, - and this is an important point – the sentence ' That finding is consistent with the lower simulated specific fluxes into the coastal ocean ' is not convincing. At the end, the Revelle factor should influence the total fluxes (and not its anthropogenic component) for which the area-based estimates indicate significantly larger negative sinks than in the global ocean (fig.5), i.e. the opposite of the anthropogenic component fluxes.

We insist that the Revelle factor does affect the anthropogenic  $CO_2$  flux (e.g., Sabine, 2004, Nature). This fact can be demonstrated with simple equilibrium calculations. For example, using CO2SYS-Matlab, if we increase the  $xCO_2$  from 280 to 400 ppm in equilibrium with two surface water masses at 2°C and 20°C each having a total alkalinity of 2300 ueq kg<sup>-1</sup>, the corresponding increases in DIC<sub>ant</sub> are 55 and 73 umol kg<sup>-1</sup> (while the Revelle factor increases by 2.2 and 1.3, respectively). Clearly the Revelle factor influences anthropogenic carbon uptake.

In the criticized sentence, we added "anthropogenic carbon" so that its revision is as follows: "That finding is consistent with the lower simulated specific fluxes of anthropogenic carbon into the coastal ocean". The differences between coastal and global ocean highlighted by the referee in Figure 5 are mainly due to natural fluxes.

Sabine et al. (2004) http://dx.doi.org/10.1126/science.1097403

Page 13 lines 23-24: The chemical factors are presented as independent of the physical factors controlling the air-sea  $CO_2$  exchange. However, based on the model construct, I feel that the higher Revelle factor for the coastal ocean precisely results from the physics of the coastal zone, with a progressive accumulation of DIC due to weaker cross-shelf export than  $CO_2$  air-sea exchange.

At the beginning of the historical simulation, the Revelle factor is already 16% larger on average, for the coastal ocean relative to the global ocean. And at the end of the simulation it remains about the same, 17% larger. We agree though that cross-shelf transport is inadequate to allow the coastal ocean to take up as much  $CO_2$  per unit area as for the global ocean average.

Figure 8: I assume that fluxes refer to total anthropogenic fluxes, i.e. organic plus inorganic carbon – please clarify.

Yes, the flux in Fig. 8 refers to anthropogenic flux. And that corresponds only to an inorganic flux. The model does not account for the anthropogenic perturbation to the organic carbon pool (see general reply).

Spelling

Page 1, Line 10-11: rephrase - this sentence is odd

#### Initial sentence:

Yet only 4.5% of that  $(0.10 \text{ Pg C yr}^{-1})$  is absorbed by the global coastal ocean, i.e., less than its 7.5% proportion of the global ocean surface area.

#### New sentence:

Yet only 0.1 Pg C yr<sup>-1</sup> of that is absorbed by the global coastal ocean. That represents 4.5% of the anthropogenic carbon uptake of the global ocean, less than the 7.5% proportion of coastal-to-global ocean surface areas.

Page 2 line 4: remove 2nd 'relative'

#### Done.

Page 4 line 25: not sure that 'model' can be used as a verb

In the revised manuscript, we replaced "explicitly models" with "explicitly accounts for".

Page 10 line 19: remove 'that'

#### OK.

Page 14 line 14: remove 'of'

Line 13: We point out a missing "carbon" after the word "anthropogenic". It will be corrected in the revised manuscript.

#### "offshore transport of carbon" is replaced by "offshore carbon transport".

Page 14 line 24-25: parenthesis wrongly placed

#### Fixed.

#### • <u>Referee #2 comments</u>

#### 1 Summary

Bourgeois et al. use an eddy-permitting global ocean biogeochemistry model to investigate the relative contribution of the ocean margins to the uptake of anthropogenic  $CO_2$  from the atmosphere. They find that these regions take up a disproportionally low amount of anthropogenic  $CO_2$ , i.e., only 4.5% relative to this regions areal contribution of 7.5%. The authors suggest that it is the limited degree to which the anthropogenic  $CO_2$  is transported and mixed offshore that leads to this low uptake. These results are in stark contrast to earlier studies based primarily on a few point observations that suggested a very high coastal ocean uptake of  $CO_2$  from the atmosphere.

#### Thanks for this nice feedback.

#### 2 Evaluation

Understanding and predicting the future evolution of the oceanic sink for atmospheric  $CO_2$  is of paramount importance for determining how much  $CO_2$  we can emit in the coming decades without exceeding any climate target. While our confidence in the net exchange fluxes of  $CO_2$  over the open ocean has increased substantially in recent years thanks to better observations and novel methods to interpret these data, our ability to constrain the fluxes along the continental margins has not increased commensurably. The existing observational constraints are still relatively weak and associated with sizeable uncertainties. By far the most extensive and detailed assessment to date by Laruelle et al. suggested a relatively small global net uptake of atmospheric  $CO_2$ , but their data-based approach did not permit them to separate this net flux into its "natural" and "anthropogenic" components.

Thus the study by Bourgeois et al. is much welcomed as it brings a consistent global perspective to the problem. Their model-based approach is far from perfect, owing to many issues ranging from its still coarse resolution to the limited of consideration of several processes that are of relevance in coastal systems (e.g., limited consideration of benthic processes), but by using - for the first time - an eddy permitting 3D model, it is a big step up from previous model based approaches that used highly simplified models. I particularly like that the authors spend a considerable amount of effort in order to assess their model's performance against observations and that they run two parallel simulations in order to determine the anthropogenic  $CO_2$  uptake explicitly. The paper is overall well written, adequately illustrated and referenced. The discussion is generally thorough and the conclusions supported by the provided evidence including an appropriate consideration of the caveats. All in all, this is a very good study that is well suited for publication in Biogeosciences. However, before giving the green light, I would like the authors to consider my concerns regarding important model shortcomings.

• Model drift: The model's drift is quite substantial, and I am not entirely convinced that the authors have fully considered the implications when discussing their results. I think the drift is large enough that it cannot be ignored.

#### Please see our general response.

• Reflective bottom boundary: The model's lower boundary is assumed to "reflect" any settling organic matter back into the water column in remineralized form. Thus, this model does not incorporate any delayed response of remineralization nor any other benthic remineralization process of relevance such as benthic denitrification etc. This is a rather important omission, as these processes influence coastal biogeochemistry in many shallow marginal seas. I thus recommend to address and discuss this issue in somewhat more detail and to give it better consideration.

## We agree the referee on the need to give better consideration on that point for the model-data comparison.

In the revised manuscript, we better described the version of the model used in this study: in particular, we indicated that part of the settling organic material is indeed reflected back in the water column in remineralized form, whereas part of it is buried to compensate for the riverine input.

Page 5 Line 15: We added "Those burial rates are hence dependent on the local sinking fluxes, but are set to balance inputs from rivers and atmospheric deposition at the global scale."

In the new version of PISCES (PISCES-v2, Aumont et al. GMD, 2015), to be used in subsequent studies, water-sediment interactions are considered using the meta-model of Middelburg et al. (1996), which allows computation of sediment denitrification. It also explicitly represents conservation of calcite in the sediment as a function of the saturation levels of the overlying waters.

In the revised manuscript, we discussed some shortcomings of such representation on our results, referring for example to the work of Krumins et al. (2013) and Soetaert et al. (2000).

Page 12 Line 19: We added "Furthermore, our simplified representation of sedimentary processes affects simulated total CO2 fluxes (Krumins et al., 2013; Soetaert et al., 2000). First, the model lacks an explicit representation of sedimentary processes. Thus it cannot reproduce the temporal dynamics of interactions between sediments and the overlying water column, e.g., resulting in potential delays between sediment burial and remineralization. Second, our model neglects any alkalinity source from sediment anaerobic degradation, such as denitrification and sulfate reduction of deposited organic matter. Even if not well constrained (Chen, 2002; Thomas et al., 2009; Hu and Cai, 2011; Krumins et al., 2013), this source of alkalinity could partially balance the total CO2 uptake of the coastal ocean. However, the simplified representation of these sediment processes has no direct effect on our anthropogenic FCO2 estimates."

Krumins et al. (2013) http://dx.doi.org/10.5194/bg-10-371-2013 Soetaert et al. (2000) http://dx.doi.org/10.1016/S0012-8252(00)00004-0

• Coastal-open ocean exchange: This issue is actually addressed a bit more than the other two in the current version, but I still consider it worthwhile to assess the implications of this shortcoming in more detail.

#### See general reply (1) above.

3 Recommendation

I recommend acceptance of this manuscript with minor revisions.

#### 4 Minor comments

Abstract, line 10: "absorbed by the coastal ocean": I suggest to define also in the abstract how the authors define the "coastal" ocean.

We agree on the need to explain our definition of the coastal ocean in the abstract. We added "Here we define the coastal zone as the continental shelf area, excluding the proximal zone." in the abstract of the revised manuscript.

p1, line 15: "the ocean naturally mitigates..." I am not sure why the authors use the expression "naturally" here. I suggest deleting it.

#### We agree. "Naturally" is deleted.

p5, line 5, equation 2: The authors still use a coefficient of 0.30, while there are numerous studies that have shown that this coefficient needs to be lowered in order to close the oceanic C14 budget. It's time to change, no?

Referee #2 is certainly correct. In future work, we will switch to using the revisited value of the coefficient (0.25) from Wanninkhof (2014, Limnol. Oceangr.), which is the new standard being adopted for the Ocean Model Intercomparison Project (OMIP). Here, we estimate that our 20% larger coefficient will lead to errors in anthropogenic carbon uptake of about 2% based on the study by Sarmiento et al (1992), who showed that a doubled coefficient increased anthropogenic carbon uptake by only 10%.

p5, lines 17 to 25: By neglecting the oceanic uptake of anthropogenic CO2 over the period 1750 until 1870, the authors underestimate the total anthropogenic uptake by about 10% or so. This should be taken into consideration more explicitly. I also suggest to avoid the use of the term "preindustrial" when referring to 1870.

In the revised manuscript, we clearly defined what we mean by preindustrial, prior to 1870, but that operational definition does indeed neglect small changes between 1750 and 1870.

Page 6 Line 12: We added "The preindustrial reference year is defined as 1870, thus neglecting changes in anthropogenic carbon storage in the ocean from 1750 to 1870."

p6, line 14: "as compared to the estimate of natural carbon outgassing of 0.45 Pg C yr-1". I find this comparison confusing, since the two processes are clearly distinct. The first number is the drift of the model, while the second one refers to the outgassing of river-derived carbon from the ocean. I do not think that these two numbers can be compared as done here.

# Our integrated air-sea flux of -0.33 Pg C/yr is actually comparable to the 0.45 Pg C yr<sup>-1</sup> outgassing because at equilibrium the model should indeed have the same value as the latter. That is, its delivery of riverine carbon to the ocean minus sedimentary burial is the same number.

p8, line 14: "Regionally, [the] overall patterns in the air-sea CO<sub>2</sub> flux are similar between the ..." How was the large model drift considered when making this statement? This issue is mentioned further down (line 24), but not really elaborated.

## We think that the general patterns in the $FCO_2$ would remain similar after a long spin up, except for the Southern Ocean where the drift is concentrated. Please see our general reply for more details.

p9, lines 16-17: "Correlation [..] only 0.5 for area-integrated fluxes". How important is the remapping error here? (see my comment on Table 2).

Remapping errors are particularly important for low area regions and reach 50% as maximums. Thus, following this comment and the next ones on Table 2 and Figure 6b, we agree Referee #2 that the model-data comparison using area-integrated  $FCO_2$  is not adequate with such large remapping errors. We removed this part and the 2<sup>nd</sup> panel (b) of the Figure 6 in the revised manuscript.

p11, lines 23-24 ".. carbon sink in the Amazon river plume [..] is not reproduced". I think that this also has something to do with the model's shortcoming with regard to the benthic-water column interactions.

Referee #2 offers a plausible explanation for this discrepancy. Another cause for discrepancy might be the instantaneous remineralisation of the entire land-derived DOC into DIC. We mentioned these possibilities in the revised manuscript.

Page 14 Line 15: We added "This discrepancy may be due to the modelled instantaneous remineralisation of land-derived DOC or to shortcomings in the model representation of sedimentary processes."

p14, lines 1-3: "... is transported offshore to the deeper open ocean". This deserves a little more discussion, particularly since a few lines below (e.g., lines 16-17) this issue is identified as "a critical question".

## We agree. In the revised manuscript, there is much discussion on this subject, as indicated in our general reply above.

Table 2: some of the areal discrepancies are huge. I thus think that one needs to be very careful when comparing areally integrated fluxes. In fact, this should not be done, in my opinion, if the areas differ by more than let's say 10%.

## Good point. We agree. Please see our comment below regarding Fig. 6, for which we removed the $2^{nd}$ panel (b) in the revised manuscript.

Figure 2: reduction in uptake during the 1940s. Where is this coming from? To me, this looks like the model is overly sensitive to the rate of change in atm. CO<sub>2</sub>, perhaps due to the drift.

Because the simulated ocean uptake of anthropogenic carbon is estimated from the difference of our 2 simulations, it bears only a very weak signature of climate (interannual, decadal) variability. The drop of uptake in the 1940s is hence clearly coming from the pause of atmospheric  $CO_2$  growth during that period. We have checked this simulated drop by comparing it to all CMIP5 historical simulations, based on the recent paper from Bastos et al. (2016) in *Biogeosciences Discussions* in which the authors focus on the 1940s  $CO_2$  plateau. The figure below indicates that our simulation is broadly consistent with the CMIP5 coupled simulations in terms of its response to the stall in atmospheric  $CO_2$  during the 1940s (see also Figure 4 from Bastos et al., 2016). As indicated in Bastos et al. (2016), "The anomalies in ocean  $CO_2$  uptake present multi-decadal variations which are consistent between the 16 models and are due to the ocean response to the atmospheric  $CO_2$  forcing. In particular, during the plateau of the 1940s, most models estimate lower ocean uptake because of the slow-down of the anthropogenic perturbation".

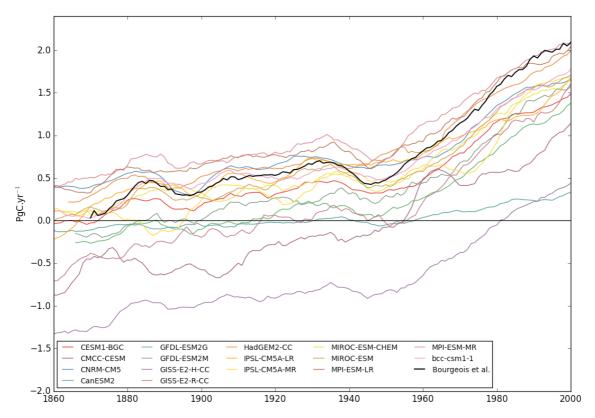


Figure 1: Ocean uptake of anthropogenic  $CO_2$  simulated by the group of CMIP5 models and comparison with our model results (black line).

Bastos, A., Ciais, P., Barichivitch, J., Bopp, L., Brovkin, V., Gasser, T., Peng, S., Pongratz, J., Viovy, N., and Trudinger, C.M. (2016). Re-evaluating the 1940s CO<sub>2</sub> plateau. Biogeosciences Discussions 1–35 (http://www.biogeosciences-discuss.net/bg-2016-171/)

Figure 6: See also my comment about Table 2. I am not sure whether it is really appropriate to compare the areally integrated fluxes when the areas are that different to begin with.

We agree. We removed Figure 6b from the revised manuscript.

## Here are some improvements done in the revised manuscript, independent from the reviewers' comments

- Add Jens Terhaar to the author list as he provided the residence-time calculation.
- Update Figs. 3d and 5 of the submitted manuscript to remove previously unnoticed errors in zonal mean calculations.
- Update Fig. 2 of the submitted manuscript by replacing time series for global ocean to time series for open ocean (see the updated figure at the end)
- Reduce file size and enhance of figure details for Figs. 3a, 3b, 3c, 3e, 3f, and 4 of the submitted manuscript.

- Figure 4 has been enriched with 2 additional panels. Panel b) is the same as Panel a) but for anthropogenic CO2 fluxes. Panel c) is a bar chart illustrating anthropogenic carbon uptake from area-integrated fluxes for each MARCATS according to MARCATS class. Figure 4's label is completed with a link to a data visualization application (http://lsce-datavisgroup.github.io/CoastalCO2Flux/)
- We provided additional details at the end of Figure 6's label: "All MARCATS regions have been used except the Black Sea, the Persian Gulf (no data estimate), and the Sea of Okhotsk (see text)". The sentence "Corrections were also applied for the Florida-Labrador delimitation" is added for information in the marked-up manuscript but is removed in the revised manuscript. See next point for details about the Florida-Labrador delimitation.
- We noticed a mistake in the shapefile delimitation for the MARCATS Florida Upwelling and Labrador Sea. The Newfoundland was linked to the Florida Upwelling whereas Newfoundland is associated to the Labrador Sea in LA13. Thus, we updated flux estimates as well as Table 2, Table 3 and Figure 1, 4, 6, and 8.
- We forgot to remove the Sea of Okhotsk estimates for the computation of R correlation coefficient. As initially stated, this observation-based estimate "is not taken into account due to the extremely poor data coverage of this region and its strong divergence with the local literature (LA14)." We updated the R correlation coefficient to 0.8, instead of 0.7 initially. This improvement is also due to the Florida-Labrador correction stated earlier.
- Replace initial wrong reference for Revelle factor in the section 2.5 by Sundquist et al. (1979)
- To enhance sentence formulation, Page 12 Line 5 is reformulated as follows: "however, they also estimate that industrialisation has recently led to a reversal in the sign of this flux (the global coastal ocean became a carbon sink) mainly due to the enhancement of NEP from increased riverine inputs."

Please find below the marked-up manuscript with additions and changes in **bold** as well as removals in **bold strikethrough**.

#### Coastal-ocean uptake of anthropogenic carbon

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Abstract. Anthropogenic changes in atmosphere-ocean and atmosphere-land  $CO_2$  fluxes have been quantified extensively, but few studies have addressed the connection between land and ocean. In this transition zone, the coastal ocean, spatial and temporal data coverage is inadequate to assess its global budget. Thus we use a global ocean biogeochemical model to assess the coastal ocean's

- 5 global inventory of anthropogenic  $CO_2$  and its spatial variability. We used an intermediate resolution, eddying version of the NEMO-PISCES model (ORCA05), varying from 20 to 50 km horizontally, i.e., coarse enough to allow multiple century-scale simulations but finer than coarse resolution models (~200 km) to better resolve coastal bathymetry and complex coastal currents. Here we define the coastal zone as the continental shelf area, excluding the proximal zone. Evaluation of the
- 10 simulated air-sea fluxes of total CO<sub>2</sub> for 45 coastal regions gave a correlation coefficient R of 0.7 0.8 when compared to observation-based estimates. Simulated global uptake of anthropogenic carbon results averaged 2.3 Pg C yr<sup>-1</sup> during 1993–2012, consistent with previous estimates. Yet only 0.1 Pg C yr<sup>-1</sup> of that is absorbed by the global coastal ocean. That represents 4.5% of the anthropogenic carbon uptake of the global ocean, less than the 7.5% proportion of coastal-
- 15 to-global ocean surface areas. Coastal uptake is weakened due to a bottleneck in offshore transport, which is inadequate to reduce the mean anthropogenic carbon concentration of coastal waters to the mean level found in the open-ocean mixed layer.

#### 1 Introduction

The ocean **naturally** mitigates climate change by absorbing atmospheric CO<sub>2</sub> produced by combustion of fossil fuels, land-use change, and cement production. During 2005–2014, the global ocean absorbed 2.6±0.5 Pg C yr<sup>-1</sup> of anthropogenic carbon, an estimated 26% of the total anthropogenic CO<sub>2</sub> emissions (Le Quéré et al., 2015). The global anthropogenic carbon budget relies on separate estimates for atmosphere, land, and ocean reservoirs. Yet it neglects what happens in the aquatic continuum between land and ocean (Cai, 2011; Regnier et al., 2013), for which there is no consensus on anthropogenic carbon uptake (Wanninkhof et al., 2013; Mackenzie et al., 2004; Bauer et al., 2013; Regnier et al., 2013; Le Quéré et al., 2015; Ciais et al., 2013).

- The land-ocean aquatic continuum includes inland waters, estuaries, and the coastal ocean, i.e., 5 the succession of active physical- biogeochemical systems that connect upland terrestrial soils to the open ocean (Regnier et al., 2013). Our focus here is on the coastal ocean, which plays an inordinately large role relative to the open ocean in terms of **relative primary productivity, export production and carbon burial**. Although the coastal ocean covers only 7-10% of the global ocean surface area, it accounts for up to 30% of oceanic primary production, 30-50% of oceanic inorganic carbon burial,
- and 80 (Gattuso et al., 1998; Longhurst et al., 1995; Walsh, 1991); moreover, the coastal ocean supplies about half of the organic carbon that is delivered to the deep open ocean (Liu et al., 2010).
   All these estimates suffer from high uncertainties as do those for coastal-ocean air-sea CO<sub>2</sub> exchange (Laruelle et al., 2014), particularly its anthropogenic component. Indeed, in addition to the effect of increasing atmospheric CO<sub>2</sub>, potential changes in coastal ocean physics (e. g.,
- 15 temperature) and biology (e.g., NEP) as well as changes in riverine input and interactions with the sediment may be of primary importance (Mackenzie et al., 2004; Hu and Cai, 2011). These changes would modify the distribution of carbon and alkalinity, and hence change the potential of the coastal ocean to absorb anthropogenic carbon.

To date, few studies have distinguished anthropogenic carbon uptake by the global coastal ocean.
Estimating air-sea fluxes of anthropogenic CO<sub>2</sub> in the coastal ocean would require multidecadal time-series of coastal CO<sub>2</sub> observations in order to extract an anthropogenic signal from the strong coastal natural variability. Such time-series are still rare and probably not long enough. To our knowledge, the only available equivalent time-series are the Ishii et al. (2011) 1994-2008 time series along 137°E on Japanese coasts and the Astor et al. (2013) 1996-2008
time-series at the CARIACO station on Venezuelan coasts. Therefore, estimates of anthropogenic series and the coastal coast

- pogenic carbon uptake by the global coastal ocean rely mainly on modelling, extrapolations from the open-ocean and/or closing- or/ balanced- budget approaches. An early modelling approach was proposed by Andersson and Mackenzie (2004) and Mackenzie et al. (2004). They used a 2-box model (Shallow-water Ocean Carbonate Model, SOCM) that separated the coastal
- 30 ocean into surface waters and sediment pore waters They estimated that the preindustrial coastal ocean was a source of CO<sub>2</sub> to the atmosphere and had recently or will switched to a CO<sub>2</sub> sink. This source-to-sink switch is mainly caused by a shift in net ecosystem production (NEP) due to increased anthropogenic nutrient inputs (Andersson and Mackenzie, 2004; Mackenzie et al., 2004). Another proposed mechanism is simply linked to the anthropogenic increase in atmo-
- 35 spheric CO<sub>2</sub>, considering constant NEP (Bauer et al., 2013). The difference between the simu-

lated air-sea  $CO_2$  fluxes from the SOCM model for years 1700 and 2000 suggests that in 2000 the coastal ocean absorbed 0.17 Pg C yr<sup>-1</sup> of anthropogenic carbon from the atmosphere (Borges et al., 2005). As for extrapolation, Wanninkhof et al. (2013) used coarse-resolution global-ocean models and observations and estimated a similar uptake of 0.18 Pg C yr<sup>-1</sup> by extrapolating open-ocean air-

5 sea fluxes of anthropogenic  $CO_2$  into the coastal zone . Finally, citeLiu2010 combined estimates from the same SOCM model for the preindustrial coastal zone with observational estimates of the contemporary flux to deduce a corresponding anthropogenic carbon uptake of 0.5  $Pg C yr^{-1}$  for the 1990s.

In addition, there exist 3-D regional circulation-biogeochemistry-ecosystem models that have been

- 10 used to study other aspects of coastal ocean carbon cycling as summarized by Hofmann et al. (2011). Typically, such models have been implemented in regions where sufficient measurements are available for model validation, e.g., the Middle Atlantic Bight (eastern U.S. coast) (Fennel et al., 2008; Fennel, 2010), the California Current System (Fiechter et al., 2014; Turi et al., 2014; Lachkar and Gruber, 2013), and the European shelf seas (Artioli et al., 2014; Phelps et al., 2014; Wakelin et al.,
- 2012; Allen et al., 2001; Cossarini et al., 2015; Prowe et al., 2009). Because of their limited regional domains, such models are typically able to make simulations with horizontal resolutions of 10 km or less, which remains a challenge for global circulation-biogeochemical models. The reduced computational requirements of regional models also allows biogeochemistry and ecosystem components to be more complex. Unfortunately, joining together a network of regional models to allow efficient
  simulations that cover all parts of the global coastal ocean remains a technical challenge (Holt et al., 2009).

The alternative of using a global model is computationally more challenging because few of them have adequate resolution to properly simulate many critical coastal-ocean processes (Griffies et al., 2010; Holt et al., 2009). Coarse-resolution global models fail to adequately resolve the coastal

- 25 bathymetry, which substantially alters coastal ocean circulation (Fiechter et al., 2014) as well as mesoscale dynamics, upwelling, and coastal currents, all of which are thought to strongly affect the variability of air-sea CO<sub>2</sub> fluxes along ocean margins (Borges, 2005; Lachkar et al., 2007; Kelley et al., 1971). Global models also typically lack a benthic component, i.e., early diagenesis in sediments, that in some regions is likely to affect simulated coastal ocean biogeochemistry of overlying
- 30 waters. Moreover input of carbon and nutrients from rivers and groundwater is usually lacking. And even in models such as ours where that input is imposed as boundary conditions (Aumont et al., 2015), temporal variability and trends are neglected (Bauer et al., 2013; Cotrim da Cunha et al., 2007).

Nonetheless, coarse-resolution models are no longer the state of the art. Recently, there have been improvements in spatial resolution of global ocean models and the spatiotemporal resolution of surface forcing fields (Brodeau et al., 2010), thereby improving the representation of bathymetry and ocean processes in the highly variable coastal zone (Capet, 2004; Hofmann et al., 2011; McKiver et al., 2014). In any case, models currently provide the only means to estimate coastal uptake of anthropogenic carbon due to the lack of data-based estimates.

- <sup>5</sup> Here our aim is to estimate the air-to-sea flux of anthropogenic  $CO_2$  into the coastal ocean and how it varies from region to region across the globe. We focus solely on the geochemical effect of anthropogenic  $CO_2$  addition from the atmosphere to the ocean and neglect the role of varying river input and interactions with the sediment, as well as the feedback from a changing climate. To do so, we rely on an eddying version of the global NEMO circulation model (Madec, 2008),
- 10 which also includes the LIM2 sea-ice model and is coupled to the PISCES biogeochemical model (Aumont and Bopp, 2006). More precisely, we use the ORCA05 eddy-admitting resolution, which ranges from 0.2° to 0.5° (i.e., 20 to 50 km). Although this resolution does not fully resolve coastal ocean bathymetry and dynamics, it does provide a first step into the eddying regime and a starting point upon which to compare future studies that will model the coastal ocean, globally, at higher
- 15 resolution.

#### 2 Methods

#### 2.1 Coupled physical-biogeochemical model

For this study, we use version 3.2 of the ocean model known as NEMO (Nucleus for European Modelling of the Ocean), which includes (1) the primitive equation model Ocean Parallélisé (OPA,

- 20 Madec (2008)), (2) the dynamic-thermodynamic Louvain-La-Neuve sea-ice model (LIM, Fichefet and Morales Maqueda (1997)), and (3) the Tracer in the Ocean Paradigm (TOP), a passive tracer module that in this case is connected to version 1 of the ocean biogeochemical model PISCES (Pelagic Interaction Scheme for Carbon and Ecosystem Studies) (Aumont and Bopp, 2006). For the NEMO model, we use a global-scale configuration from the DRAKKAR community (see Barnier)
- et al. (2006) and Timmermann et al. (2005)). Namely, we use the ORCA05 global configuration, which possesses a curvilinear, tri-polar grid with a horizontal resolution that ranges between 0.2° near the North Pole to 0.5° at the equator (Fig. 1). Vertically, ORCA05 is discretized into 46 levels with thicknesses that range from 6 m at the surface to 250 m for the deepest ocean level (centered at 5625 m). Model bathymetry is computed from the 2' bathymetry file ETOPO2 from the National
- 30 Geophysical Data Center. The numerical characteristics of our ORCA05 configuration follow the lead of Barnier et al. (2006) for the ORCA025 configuration with resolution-dependent modifications for the horizontal eddy diffusivity for tracers modified to 600 m<sup>2</sup> s<sup>-1</sup> and horizontal eddy viscosity fixed to  $-4 \times 10^{11} \text{ m}^2 \text{ s}^{-1}$ . To simulate the advective transport driven by geostrophic eddies,

our ORCA05 simulation uses the eddy parameterization scheme of Gent and McWilliams (1990) applied with an eddy diffusion coefficient of  $1000 \text{ m}^2 \text{s}^{-1}$ .

The biogeochemical model PISCES includes four plankton functional types: 2 phytoplankton (nanophytoplankton and diatoms) and 2 zooplankton (micro- and meso-zooplankton). PISCES also uses a mixed quota-Monod approach where (1) phytoplankton growth is limited by 5 nutrients (nitrate, ammonium, phosphate iron and silicate) following Monod (1949) and (2) elemental ratios of Fe, Si and Chl to C are prognostic variables based on the external concentrations of the limiting nutrients. In addition PISCES assumes a fixed C:N:P Redfield ratio set to 122 : 16 : 1 from Takahashi et al. (1985) for both living and non-living pools. Similar to Geider et al. (1998), the phytoplankton

- 10 Chl:C ratio in PISCES varies with photoadaptation. Furthermore, PISCES includes nonliving pools, namely a pool of semi-labile dissolved organic matter and two size classes of particulate organic matter. PISCES also explicitly accounts for biogenic silica and calcite particles. In PISCES, the sediment-water interface is treated as a reflective boundary condition where mass fluxes from particles are remineralized instantaneoulsy, except that small proportions of particle fluxes of organic
- 15 matter, calcite, and biogenic silica escape the system through burial. Those burial rates are hence dependent on the local sinking fluxes, but are set to balance inputs from rivers and atmospheric deposition at the global scale. Thus global budgets of alkalinity and nutrients are balanced. For further details, we refer readers to Aumont and Bopp (2006).

To simulate carbon chemistry and air-sea CO<sub>2</sub> fluxes, the model follows the protocol from phase
20 2 of the Ocean-Carbon Cycle Model Intercomparison Project (OCMIP, Najjar and Orr (1999)) protocol. The sea-to-air CO<sub>2</sub> flux FCO<sub>2</sub> is computed using the following equation:

$$FCO_2 = \alpha k \Delta p CO_2 \tag{1}$$

where α is the solubility of CO<sub>2</sub> computed from Weiss (1974) and Δ*pCO*<sub>2</sub> is the difference between the partial pressures of sea-surface and atmospheric CO<sub>2</sub>. Thus FCO<sub>2</sub> is positive when CO<sub>2</sub>
25 is transferred from the ocean to the atmosphere. The piston velocity k is based on equation (3) of Wanninkhof (1992) as:

$$k = 0.30 \ u_w^2 \sqrt{\frac{660}{Sc}} (1 - f_{ice}) \tag{2}$$

where  $u_w$  is the wind speed at 10 m, Sc is the CO<sub>2</sub> Schmidt number, and  $f_{ice}$  is the ice fraction.

#### 2.2 Simulations

30 The dynamic model was started from rest and spun up for 50 years. Initial conditions for temperature and salinity are as described by Barnier et al. (2006). Initial biogeochemical fields of nitrate, phosphate, oxygen and silicate are from the 2001 World Ocean Atlas (Conkright et al., 2002), whereas preindustrial dissolved inorganic carbon (DIC) and total alkalinity (Alk) come from the GLODAP gridded product (Key et al., 2004). Conversely, because data for iron and dissolved organic carbon (DOC) are more limited, both those fields were initialized with model output from a 3000-year spinup simulation of a global 2° configuration of the same NEMO-PISCES model (Aumont and Bopp,

5 2006). All other biogeochemical tracers have much shorter time-scales; hence, they were initialized to globally uniform constants.

After the 50-year spin up, we launched 2 parallel simulations: the first was a historical simulation run during 1870 to 2012 (143 years), and forced with a spatially uniform and temporally increasing atmospheric mole fraction of CO<sub>2</sub> (from which PISCES computes atmospheric  $pCO_2^{atm}$  following

- 10 OCMIP2) reconstructed from ice-core and atmospheric records (Le Quéré et al., 2014); the second simulation is a parallel control run, where the 143-year simulation is identical except that it is forced with the preindustrial level of atmospheric mole fraction of CO<sub>2</sub> (287 ppm, constant in time). The preindustrial reference year is defined as 1870, thus neglecting changes in anthropogenic carbon storage in the ocean from 1750 to 1870. The FCO<sub>2</sub> computed with the historical simulation is
- 15 for total carbon (total  $FCO_2$ ), whereas that from the control simulation is for natural carbon (natural  $FCO_2$ ). The corresponding anthropogenic  $FCO_2$  is computed as the total minus natural  $FCO_2$ .

All simulations were forced identically, with atmospheric fields from the DRAKKAR Forcing Set (DFS, Brodeau et al. (2010)). These fields include zonal and meridional components of 10-m winds, 2-m air humidity, 2-m air temperature, downward shortwave and longwave radiation at the

20 sea surface, and precipitation. More specifically the NEMO-PISCES model is forced with version 4.2 of this forcing (DFS4.2, based on the ERA40 reanalysis) over 1958–2001, and that is followed by forcing from version 4.4 (DFS4.4) over 2002 to 2012. For the 1870–1957 period, where atmospheric reanalyses are unavailable, we repeatedly cycled the 1958-2007 DFS4.2 forcing.

Boundary conditions are also needed for biogeochemical tracers, i.e., besides the atmospheric-25 CO<sub>2</sub> connection mentioned already. The model's lateral input from river discharge of DIC and DOC are taken from the annual estimates of the Global Erosion Model (Ludwig et al., 1996), constant in time. The DOC from river discharge is assumed to be labile and is directly converted to DIC upon its delivery to the ocean. Inputs of dissolved iron (Fe), nitrate ( $NO_3^{2-}$ ), phosphate ( $PO_4^{3-}$ ), and silicate (SiO<sub>2</sub>) are computed from the sum of DIC and DOC river input using a constant set of ratios for

30 C:N:P:Si:Fe, namely 320 : 16 : 1 : 53.3 : 3.64 x 10<sup>-3</sup>, as computed from Meybeck (1982) for C:N, from Takahashi et al. (1985) for N:P, from de Baar and de Jong (2001) for Fe:C, and from Treguer et al. (1995) for Si:C. River discharge assumes no seasonal variation. Atmospheric deposition of iron comes from Tegen and Fung (1995).

Here, we use the conventional definition of anthropogenic carbon in the ocean used by previous global-ocean model studies (OCMIP, http://ocmip5.ipsl.jussieu.fr/OCMIP/ and e.g., Bopp et al. (2015)), namely that anthropogenic carbon comes only from the direct geochemical effect of increasing atmospheric  $CO_2$  and its subsequent invasion into the ocean. By definition, this anthropogenic  $FCO_2$  does not include any effect from potential changes in ocean physics or biology. In the model, there are no changes nor variability in riverine delivery of carbon and

5 nutrients, and anthropogenic carbon is not buried in sediments.

Following the 50-year spin up and 143-year control simulation, the simulation remains far from equilibrium. Its global natural carbon flux is  $-0.33 \pm 0.3 \text{ Pg C yr}^{-1}$  (corresponding to CO<sub>2</sub> uptake by the ocean) during the last 10 years of the control simulation (2003-2012), as compared to the estimate of natural carbon outgassing of 0.45 Pg C yr<sup>-1</sup> by Jacobson et al. (2007). That difference is

- 10 partly due to the strategy for our simulations, which were initialized with data and spun up for only 50 years because of the computational constraints to make higher resolution simulations (ORCA05). At lower resolution (ORCA2), after a spin-up of 3000 years, there is 0.26 Pg C yr<sup>-1</sup> greater globally integrated sea-to-air flux, relative to results after only a 50-year spin up. Nearly all of that enhanced sea-to-air CO<sub>2</sub> flux due to the longer spin up comes from the Southern Ocean. Anthropogenic FCO<sub>2</sub>
- 15 estimates are expected to be influenced very little by model drift because of the way anthropogenic carbon is defined, i.e., drift affects both natural carbon and total carbon in the same way.

#### 2.3 Defining the global coastal ocean

To sample the global coastal ocean area, the model grid cells were selected following the Margins and Catchments Segmentation (MARCATS) of Laruelle et al. (2013), hereafter LA13. The outer limit of the coastal ocean is defined as the maximum slope at the shelf break, while the inner limit is taken as the coastline, thus excluding the proximal zone of the coastal ocean (Fig. 1). Hence, only the continental shelf area is taken into account. The MARCATS segmentation divides the global coastal ocean into 45 regional units (Table 2). The limits of each of theses units delineate areas that

- 25 present roughly homogenous oceanic features such as coastal currents or the boundaries of marginal seas. Following the Liu et al. (2010) classification of the continental shelf seas, LA13 aggregated the 45 units into 7 classes with similar physical and oceanographic large-scale characteristics such as the Eastern Boundary Currents and the Polar Margins. The high-resolution Geographical Information System (GIS) file describing the MARCATS segmentation from LA13 was regridded using the QGIS
- 30 software (QGIS Development Team, 2015) on the ORCA05 model grid in order to sample the model results on its own grid. This regridding technique implies some modifications to the regions initially described in LA13. In the model, the global coastal ocean has a total surface area of 27.0 x 10<sup>6</sup> km<sup>2</sup>, which is 8% less than the original value from Laruelle et al. (2014). Here, the model's total coastal

ocean surface area represents 7.5% of the total area of the global ocean. Subsequently we refer to the individual MARCATS regions using the terminology of LA13.

#### 2.4 Evaluation dataset

To evaluate the total  $FCO_2$  simulated by the model (historical simulation), we compare it to the

- 5 database from Laruelle et al. (2014), hereafter LA14, which provides observation-based estimates for that flux over the MARCATS regions. This database was constructed by aggregating  $3 \times 10^6$ coastal sea-surface *p*CO<sub>2</sub> measurements collected during 1990 to 2011 and included in the Surface Ocean CO<sub>2</sub> Atlas version 2.0 (SOCAT v2.0, Pfeil et al. (2013); Bakker et al. (2014)). These measurements represent about 30% of the SOCAT v2.0 dataset. To compute the flux, LA14 also relied
- 10 on wind speeds from the multiplatform CCMP wind-speed database (Atlas et al., 2011), atmospheric CO<sub>2</sub> from GLOBALVIEW-CO2 (2012), and the flux parameterization from Wanninkhof (1992) as modified by Takahashi et al. (2009). As sensitivity tests, LA14 also used the flux parameterizations from Ho et al. (2006) as well as the original formulation from Wanninkhof (1992).

Thus LA14 computed mean annual FCO<sub>2</sub> estimates for 42 of the 45 MARCATS regions defined in

- 15 LA13. The remaining MARCATS areas (12:Hudson Bay, 21:Black Sea and 29:Persian Gulf) are devoid of observations in the SOCAT database and were neglected. For the remaining regions, because of the large heterogeneity in both the spatial and temporal coverage of ocean *p*CO<sub>2</sub> observations, the uncertainties for each the MARCATS FCO<sub>2</sub> estimates from LA14 vary greatly. For example, only 28% of the sub-units of MARCATS regions used in LA14 have an estimate for FCO<sub>2</sub> uncertainty
- of less than 0.25 mol C m<sup>-2</sup> yr<sup>-1</sup>. The data-based FCO<sub>2</sub> estimate for the Sea of Okhotsk is not taken into account due to the extremely poor data coverage of this region and its strong divergence with the local literature (LA14). Here, we do not evaluate the simulated annual cycle of flux of total carbon because few MARCATS regions provide adequate temporal coverage. Finally, LA14 is the first and only study to provide coastal-ocean observation-based FCO<sub>2</sub> estimates at global scale
- **25** taking into account for the reduction in  $FCO_2$  due to sea-ice cover along coasts; hence it is directly comparable to our model results.

Besides the coastal data-based estimates of  $FCO_2$  from LA14, we also compare our model results to those for the open ocean from Takahashi et al. (2009) and Landschützer et al. (2014). Both the global and coastal observational estimates are compared to the average modeled  $FCO_2$  over the last

30 20 years (1993–2012) of the historical simulation. For the coastal comparison, simulated total FCO<sub>2</sub> are spatially averaged over each MARCATS regions. In addition, the model's uncertainty, **computed as the interannual variability over 1993–2012**, is compared to uncertainties in the observational estimates , **computed as the standard deviation between flux parameterizations from Wanninkhof** (1992) as modified by Takahashi et al. (2009), Ho et al. (2006) and Wanninkhof (1992).

#### 2.5 Revelle factor calculation

To assess how the capacity of the coastal ocean to absorb anthropogenic carbon differs from openocean surface waters, we computed the Revelle factor ( $R_f$ , **Sundquist et al.** (1979)) using the CO2SYS MATLAB algorithm (Van Heuven et al., 2011). CO2SYS was used using the simulated sea-surface temperature, salinity, alkalinity, and DIC for model years 1993–2012 with the total pH

scale, the  $K_1$  and  $K_2$  constants from Lueker et al. (2000), the  $K_{SO_4}$  constant from Dickson (1990) and the formulation of the borate-to-salinity ratio from Uppström (1974).

#### 2.6 Residence time

To compute water residence time in each MARCATS region, we divided the volume of each region by the integrated outflow of water from 5-day mean current velocities at coastal boundaries from 2011.

#### **3** Results

5

#### 3.1 Global ocean fluxes

The simulated global-ocean uptake of anthropogenic carbon increases roughly linearly from 1950 to
2012, reaching an average of 2.3 Pg C yr<sup>-1</sup> during 1993–2012. That is comparable to the estimate from the fifth assessment report of the IPCC (Ciais et al., 2013) of 2.3±0.7 Pg C yr<sup>-1</sup> for 2000–2009 (Fig. 2).

Regionally, overall patterns in the total  $FCO_2$  are similar between the model and data-based estimates from Landschützer et al. (2014) and Takahashi et al. (2009) (Fig. 3). Carbon is lost from the

- 20 ocean in the equatorial band and in coastal upwelling regions, while it is gained by the ocean in the northern high latitudes. Quantitative comparison of the annual-mean map from the model with that from the Takahashi et al. (2009) observation-based database gives a root mean square error (RMSE) of 0.73 mol C m<sup>-2</sup> yr<sup>-1</sup> and a correlation coefficient R of 0.80; likewise, comparison with the Landschützer et al. (2014) observational-based database gives a similar RMSE (0.70 mol C m<sup>-2</sup> yr<sup>-1</sup>)
- and R (0.81). Integrating over latitudinal bands, (Table 1), the model overestimates carbon uptake for the 90°S-30°S region where it absorbs 1.50  $PgCyr^{-1}$  of total carbon versus 0.73-0.77  $PgCyr^{-1}$ from Takahashi et al. (2009) and Landschützer et al. (2014) observational databases. This may be a signature of the fact that the model simulation is still far from equilibrium (see section 2.2 paragraph 5 for details). The model also underestimates outgassing in the tropical band, where it releases
- 30  $0.13 \text{ Pg C yr}^{-1}$  vs. 0.13-0.20  $\text{Pg C yr}^{-1}$  for the 2 data-based estimates. Further north in the 30°N-

90°N band the model takes up 0.93  $Pg Cyr^{-1}$  vs. 1.53–1.59  $Pg Cyr^{-1}$  for Takahashi et al. (2009) and Landschützer et al. (2014).

#### 3.2 Coastal ocean fluxes

#### 3.2.1 Total CO<sub>2</sub>

- 5 The simulated uptake of total carbon by the coastal ocean averages  $267 \text{ Tg C yr}^{-1}$  during the 1993– 2012. Most of the 45 MARCATS regions act as carbon sinks; together, they absorb 283 Tg C yr<sup>-1</sup>. The largest uptake is 3.4 mol C m<sup>-2</sup> yr<sup>-1</sup> in the South Greenland region. Few MARCATS regions act as carbon sources to the atmosphere (Table 2 and Fig. 4.a), i.e., only 14% of the global coastalocean surface area, together losing 16 Tg C of carbon to the atmosphere every year. The mean
- 10 annual carbon loss per square meter in these MARCATS regions is usually relatively weak, less than 1.5 mol C m<sup>-2</sup> yr<sup>-1</sup>). When grouped into MARCATS classes (see Table 3), all classes are carbon sinks, absorbing from 0.06 to 1.65 mol C m<sup>-2</sup> yr<sup>-1</sup>. By class, the largest specific fluxes occur in the Western Boundary Current regions and the Subpolar Margins, which absorb 1.65 and 1.61 mol C m<sup>-2</sup> yr<sup>-1</sup>, respectively. More generally, the tropical MARCATS regions act as **weak**
- 15 carbon sources and the mid-to-high latitude regions act as **strong** carbon sinks (Fig. 4.a). The same trend is also apparent in the zonal-mean distribution (Fig. 5).

A comparison of the simulated vs. observed  $FCO_2$  estimates for each MARCATS region is reported in Table 2 and on Fig. 6. The Pearson correlation coefficient R is **0.7 0.8** for specific fluxes **, but only 0.5 for area-integrated fluxes**. In the model, 79% of the MARCATS regions act as carbon

- 20 sinks, whereas that proportion is 64% for LA14. After aggregating the specific flux estimates into the different MARCATS classes (Table 3 and Fig. 7), the correlation coefficient R increases to 0.9. Generally, our model results tend to simulate larger sinks and weaker sources than observed (i.e. 76% of the specific simulated fluxes of total carbon have lower relative values than the databased estimates). For some MARCATS classes, even the sign of the simulated flux differs from
- 25 the data-based estimates, e.g., for the Indian Margins and the Eastern Boundary Currents. The latter class contains two regions (Moroccan and S-W Africa Upwelling) having the worst overall agreement. **Otherwise**, in the Arctic polar regions, the simulated uptake is too low, with 52 Tg C yr<sup>-1</sup> from the model vs. 86 Tg C yr<sup>-1</sup> from LA14.

#### 3.2.2 Anthropogenic CO<sub>2</sub>

30 The anthropogenic FCO<sub>2</sub> is computed as the difference between the total flux (historical simulation) and natural flux (control simulation). When integrated over the global coastal ocean, the mean anthropogenic flux during 1993–2012 is  $0.10\pm0.01$  Pg C yr<sup>-1</sup>. That amounts to 4.5% of the simulated

global anthropogenic carbon uptake, substantially less than the 7.5% proportion of the coastal-toglobal ocean surface areas. During 1950–2000, the uptake of anthropogenic carbon by the coastal ocean grows essentially linearly as it does for the global ocean. That is, it grows at a nearly constant rate of 0.0015 Pg C yr<sup>-2</sup>, which is 4.4% of the rate for the global ocean increase in anthropogenic earbon uptake over the same period (Fig. 2).

5 carbon uptake over the same period (Fig. 2).

All MARCATS regions absorb anthropogenic carbon at rates ranging from 0.01 mol C m<sup>-2</sup> yr<sup>-1</sup> for the Baltic Sea to 0.86 mol C m<sup>-2</sup> yr<sup>-1</sup> for the South Greenland region (Table 2 and Fig. 4.b). By class, the strongest specific fluxes of anthropogenic carbon into the ocean occur in the boundary current regions, namely the EBC and WBC, with 0.42 and 0.48 mol C m<sup>-2</sup> yr<sup>-1</sup>, respectively.

10 Conversely, the weakest anthropogenic carbon uptake occurs in the Tropical Margins and the Indian margins with 0.22 and 0.24 mol C m<sup>-2</sup> yr<sup>-1</sup>, respectively. But specific fluxes can be misleading. Although the Polar and Subpolar margins do not have the highest specific fluxes, their integrated uptake of anthropogenic carbon is large because of their large surface areas (Fig. 4.b and 4.c). Together they absorb 46 Tg C yr<sup>-1</sup>, which is 45% of total uptake of anthropogenic carbon by the global coastal

15 ocean.

These results emphasize that there is no link between anthropogenic and total carbon fluxes when comparing patterns between regions. For example, even though the EBC and WBC regions are **the most efficient regions in anthropogenic carbon uptake** (both above 0.4 mol C m<sup>-2</sup> yr<sup>-1</sup>), their behavior differs greatly in terms of the flux of total carbon, i.e., -1.65 versus -0.12 mol C m<sup>-2</sup> yr<sup>-1</sup>,

20 respectively (Fig. 7). The same lack of correlation between anthropogenic and total flux patterns is even clearer in the zonal mean distributions (Fig. 5). For instance, the specific fluxes of anthropogenic carbon into the coastal ocean between 55°S and 90°N are nearly uniform, remaining near -0.5 mol C m<sup>-2</sup> yr<sup>-1</sup>; conversely, the total carbon fluxes **that** vary greatly, between -2 to +0.5 mol C m<sup>-2</sup> yr<sup>-1</sup>. Theses variations in the total carbon flux are dictated by variations in the natural carbon flux (Fig. 5).

#### 4 Discussion

#### 4.1 Comparison with previous coastal estimates

#### 4.1.1 Total flux

Our mean simulated uptake of total carbon by the global coastal ocean during the 1993–2012 is

0.27±0.07 Pg C yr<sup>-1</sup>, which falls within the range of previous data-based estimates of 0.2–0.4 Pg C yr<sup>-1</sup> (Borges et al., 2005; Cai et al., 2006; Chen and Borges, 2009; Laruelle et al., 2010; Cai, 2011; Chen et al., 2013; Laruelle et al., 2014). Out of those, estimates provided since 2011 gather closer to the

lower limit, e.g., the estimate of  $0.2 \text{ Pg C yr}^{-1}$  from LA14, as is also the case for our model-based estimate. Some aspects of the LA14 data-based approach are shared by our model-based approach, i.e., the same reference period, essentially the same definition of the coastal ocean and the same correction for the effect of sea-ice cover on FCO<sub>2</sub>. LA14 is the first observation-based study to take

#### 5 into account this sea-ice effect for coastal-ocean FCO<sub>2</sub> estimates at global scale.

Using a box model, Andersson and Mackenzie (2004) and Mackenzie et al. (2004) estimated the global coastal ocean acted as a carbon source to the atmosphere prior to industrialisation; however, they also estimate that industrialisation has **recently** led **or will lead** to a reversal in the sign of this flux (the global coastal ocean **will become became** a carbon sink) **sometime between 1950 and** 

- 10 2100 mainly due to the enhancement of NEP from increased riverine inputs. In contrast, our model simulations indicate that the preindustrial coastal ocean was already a carbon sink, and that that sink has strengthened over the industrial period. This discrepancy appears to be explained by different definitions of the coastal ocean. Both the box model and our 3-D model include the distal coastal zone, but only the box model includes the proximal coastal zone (bays, estuaries, deltas,
- 15 lagoons, **salt marshes, mangroves, and banks**). That proximal zone is known generally as a strong source of carbon to the atmosphere (Rabouille et al., 2001).

The model representation of riverine DOC input and its instantaneous remineralization has potential implications for our estimates of total  $FCO_2$ . In the Amazon plume for instance, we underestimate  $CO_2$  absorption because of this instantaneous addition of DIC without input of alkalinity. However this assumption has no direct implication on our anthropogenic  $FCO_2$  estimates.

Furthermore, our simplified representation of sedimentary processes affects simulated total  $CO_2$  fluxes (Krumins et al., 2013; Soetaert et al., 2000). First, the model lacks an explicit representation of sedimentary processes. Thus it cannot reproduce the temporal dynamics of

- 25 interactions between sediments and the overlying water column, e.g., resulting in potential delays between sediment burial and remineralization. Second, our model neglects any alkalinity source from sediment anaerobic degradation, such as denitrification and sulfate reduction of deposited organic matter. Even if not well constrained (Chen, 2002; Thomas et al., 2009; Hu and Cai, 2011; Krumins et al., 2013), this source of alkalinity could partially balance the to-
- 30 tal CO<sub>2</sub> uptake of the coastal ocean. However, the simplified representation of these sediment processes has no direct effect on our anthropogenic FCO<sub>2</sub> estimates.

#### 4.1.2 Anthropogenic flux

20

The strongest specific fluxes of anthropogenic carbon into the ocean occur in the boundary current regions, namely the EBC and WBC. Indeed, these regions show significant vertical and lateral mixing features such as filaments and eddies from the strong adjacent western boundary currents and upwelling from Eastern Boundary Upwelling Systems (EBUS). Those physical processes lead to deepen the mixed layer depth, export the absorbed anthropogenic carbon from shallow water to deeper water layers and export it to the adjacent open ocean.

- 5 Our estimate of the simulated anthropogenic carbon uptake of  $0.10 \text{ Pg C yr}^{-1}$  for the global coastal ocean (Fig. 9) is about half that found by Wanninkhof et al. (2013) for a similar period. The latter study estimates coastal anthropogenic CO<sub>2</sub> uptake by extrapolating specific FCO<sub>2</sub> from the adjacent open ocean into coastal areas, exploiting coarse-resolution models and data. To compare approaches, we applied the Wanninkhof et al. (2013) extrapolation method to our model out-
- 10 put; we found the same result as theirs for global coastal ocean uptake of anthropogenic  $CO_2$ (0.18 Pg C yr<sup>-1</sup>). Thus the extrapolation technique leads to an overestimate of anthropogenic  $CO_2$ uptake in the model's global coastal ocean.

Nonetheless, the Wanninkhof et al. (2013) estimate for the anthropogenic carbon uptake by the coastal ocean was used by Regnier et al. (2013) for their coastal carbon budget. That budget also

- 15 accounts for the increase in river discharge of carbon  $(0.1 \text{ Pg C yr}^{-1})$  and nutrients during the industrial era, which promotes organic carbon production, some of which is buried in the coastal zone (up to  $0.15 \text{ Pg C yr}^{-1}$ ). Unfortunately, these numbers remain particularly uncertain. Hence we have chosen to ignore them, adopting the conventional definition of anthropogenic carbon in the ocean used by previous global-ocean model studies, namely that anthropogenic carbon comes only from
- 20 the direct geochemical effect of the anthropogenic increase in atmospheric  $CO_2$  and its subsequent invasion into the ocean. The future challenge of improving estimates of changes and variability in riverine delivery of carbon and nutrient and sediment burial is critical to refine land contributions to the coastal ocean carbon budget.

Our estimate of 0.10  ${\rm Pg\,C\,yr^{-1}}$  for the anthropogenic  $FCO_2$  into the coastal ocean is 40% less

25 than the 0.17 Pg C yr<sup>-1</sup> estimated by Borges et al. (2005) from Andersson and Mackenzie (2004) and Mackenzie et al. (2004). Causes for this difference may stem from (1) the different definitions of the coastal ocean (proximal coastal zone included in the box model but not the 3-D model), (2) the different approaches (uniform coastal ocean in the box model but not in the 3-D model), and (3) the role of sediments (pore waters included in the box model but neglected in the 3-D model).

#### 30 4.2 Coastal vs. open ocean

Patterns in our simulated total FCO<sub>2</sub> in the coastal ocean generally follow those for the open ocean, with net carbon sources in the low latitudes and carbon sinks in the mid- to high-latitudes (Fig. 5). The same tendency was pointed out by Gruber (2014) when discussing the LA14 data-based fluxes. The patterns in our simulated total CO<sub>2</sub> flux are mainly driven by patterns in the natural CO<sub>2</sub> flux both in the coastal and open oceans (Fig. 5). Yet the pattern for anthropogenic  $CO_2$  flux differs greatly from that of natural  $CO_2$ , having its strongest uptake in the Southern Ocean in both the open and coastal oceans, i.e., where zonally averaged specific uptake reaches up to 1.5 mol C m<sup>-2</sup> yr<sup>-1</sup>. The bathymetry of MARCATS regions around the Antarctic continent

5 is much deeper than in the other coastal regions (500 m vs. 160 m for the global coastal ocean); this probably reduces the contrast between the coastal and open ocean in the Southern Ocean and explains the similarities of anthropogenic carbon uptake rates there.

Despite large-scale similarities between coastal and open-ocean fluxes of total carbon, some coastal regions differ substantially from those in the adjacent open ocean waters (Fig. 3.a). These local dif-

- 10 ferences are particularly apparent around coastal upwelling systems, i.e., in the Western Arabian Sea and in Eastern Boundary Upwelling Systems (EBUS), such as the Peruvian Upwelling Current, the Moroccan Upwelling, and the Southern Western Africa upwelling. Some of these coastal regions act as strong total carbon sources, with mean carbon fluxes of up to 1.44 mol C m<sup>-2</sup> yr<sup>-1</sup>, whereas surrounding open-ocean waters exhibit little FCO<sub>2</sub> (fluxes close to 0 mol C m<sup>-2</sup> yr<sup>-1</sup>). Other regions
- 15 also exhibit large coastal-open ocean contrasts, including the Tropical Western Atlantic where there is a massive loss of carbon at the location of the Amazon river discharge. However the carbon sink in the Amazon river plume reported in Lefèvre et al. (2010) is not reproduced. This discrepancy may be due to the modelled instantaneous remineralisation of land-derived DOC or to shortcomings in the model representation of sedimentary processes.
- A key finding of our model study is that the flux of anthropogenic CO<sub>2</sub> into the coastal ocean (0.10 Pg C yr<sup>-1</sup>) is half the previous estimate (Wanninkhof et al., 2013). Unlike in that study, our specific flux of anthropogenic CO<sub>2</sub> is substantially lower for the global coastal ocean than for the global open ocean (i.e., -0.31 vs. -0.54 mol C m<sup>-2</sup> yr<sup>-1</sup> for the 1993–2012 average). Although the coastal ocean surface area is 7.5% that of the global ocean, it absorbs only 4.5% of the globally integrated flux of anthropogenic carbon into the ocean.

Our estimate for coastal ocean uptake of anthropogenic carbon is ten times smaller than the  $1 \text{ Pg C yr}^{-1}$  estimate by Tsunogai et al. (1999) associated with his proposed continental shelf pump (CSP). However, Tsunogai's CSP is based on contemporary measurements and thus concerns to-tal carbon, not the anthropogenic change. That nuance is critical because contemporary estimates

- 30 of fluxes are not directly comparable to anthropogenic fluxes nor global budgets of carbon from the IPCC and the Global Carbon Project, both focused on the anthropogenic change. Unfortunately Tsunogai et al. (1999) prompted confusion by stating that their total carbon flux into the coastal ocean was equivalent to half of the global-ocean uptake of anthropogenic carbon. The same confusion prompted Thomas et al. (2004) to emphasize that the coastal ocean contributes more to the
- 35 global carbon budget than expected from its surface area.

The lower specific flux of anthropogenic  $CO_2$  into the global coastal ocean relative to the average for the open ocean could have 2 causes: (1) physical factors, e.g., if vertical mixing in the coastal ocean is relatively weak or if there is a bottleneck in the offshore transport carbon and (2) chemical factors, if coastal waters have a lower chemical capacity to absorb anthropogenic carbon (lower carbonate ion concentration, higher Revelle factor  $R_f$ ).

5

To assess how  $R_f$  differs between coastal and open-ocean surface waters, we computed it using CO2SYS from simulated sea-surface temperature, salinity, alkalinity, and DIC for model years 1993–2012. Thus we computed mean Revelle factors over of 12.5 for the global coastal ocean, 10.9 for the global ocean, 9.2 for the tropical oceans (30°S-30°N), and 12.8 for the Southern Ocean (90°S-30°S).

- 10 And these tendencies are persistent. During 1910–2012, the average coastal-ocean Revelle factor remains 15% larger than that for the open ocean. Hence average surface waters in the model's coastal ocean have a lower chemical capacity to take up anthropogenic carbon than do average surface waters of the global ocean. That finding is consistent with the lower simulated specific fluxes of anthropogenic carbon into the coastal ocean. Yet it is not only the chemical capacity that matters. For
- 15 example, despite similar chemical capacities, the specific flux of anthropogenic carbon into Southern Ocean is more than twice that of the global coastal ocean. Thus we must turn to physical factors to help explain the lower efficiency of the coastal ocean to take up anthropogenic carbon.

Out of the  $0.10 \,\mathrm{Pg}\,\mathrm{C}\,\mathrm{yr}^{-1}$  absorbed by the coastal ocean, we find that only 70% (i.e.  $0.07 \,\mathrm{Pg}\,\mathrm{C}\,\mathrm{yr}^{-1}$ ) is transferred to the open ocean (Fig. 9). Thus  $0.03 \,\mathrm{Pg}\,\mathrm{C}\,\mathrm{yr}^{-1}$  of anthropogenic carbon accumulates

- in the coastal-ocean water column during 1993–2012. That simulated accumulation is not significantly different from the estimate of  $0.05 \pm 0.05$  Pg C yr<sup>-1</sup> from Regnier et al. (2013). The accumulation in the coastal ocean is effective over the entire period (1910-2012) as the uptake of anthropogenic carbon by the global coastal ocean is always inferior to its cross-shelf export (Fig. 10). To gain insight into this cross-shelf exchange, we computed the simulated mean
- 25 water residence times for each MARCATS region (Fig. 8). Residence times for most coastal regions are of the order of a few months or less, except for Hudson Bay, the Baltic Sea and the Persian Gulf. The latter three regions are generally more confined and we expect longer residence times, although our model simulations were never designed to simulate these regions accurately. Generally, our simulated residence times are shorter than what has been published
- 30 for similarly defined coastal regions although methods differ substantially (Jickells, 1998; Men and Liu, 2014; Delhez et al., 2004). Despite these short residence times, the cross-shelf export of anthropogenic carbon is unable to keep up with the increasing air-sea flux of anthropogenic carbon (Fig. 10). This may be explained by the open-ocean waters that are imported to the coastal ocean being already charged with anthropogenic carbon, thus limiting further uptake
- 35 in the coastal zone. This accumulation rate of anthropogenic carbon in the coastal ocean contrasts

with the lower simulated proportion that remains in the mixed layer of the global ocean. Using a coarse-resolution global model, Bopp et al. (2015) showed that on average for the global ocean, only  $\sim 10\%$  of the anthropogenic carbon that crosses the air-sea interface accumulates in the seasonally-varying mixed layer. The CSP hypothesis from Tsunogai et al. (1999) assumes that much of the 1

- 5 Pg C yr<sup>-1</sup> of total carbon absorbed by the coastal ocean is exported to the deep ocean. Also assuming that the CSP operates equally in all shelf regions across the world, Yool and Fasham (2001) used coarse- resolution global model to estimate that 53% of the coastal uptake is exported to the open ocean. Yet they considered only natural carbon. Conversely, we focus purely on anthropogenic carbon. Our simulations suggest that 70% of the anthropogenic carbon absorbed by the coastal ocean
- 10 during 1993–2012 is transported offshore to the deeper open ocean.

#### 5 Conclusions

The goal of this study is to estimate the anthropogenic  $CO_2$  flux from the atmosphere to the coastal ocean, both globally and regionally, using an eddying global-ocean model, making 143-year simulations forced by atmospheric reanalysis data and atmospheric  $CO_2$ . We first evaluate the simulated

- 15 air-sea fluxes of total CO<sub>2</sub> for 45 coastal regions and find a correlation coefficient R of 0.8 when compared to observation-based estimates. Then we estimate the average simulated anthropogenic carbon uptake by the global coastal ocean over 1993–2012 to  $0.10 \pm 0.01 \text{ Pg C yr}^{-1}$ , equivalent to 4.5% of global-ocean uptake of anthropogenic CO<sub>2</sub>, an amount less than expected based on the surface area of the global coastal ocean (7.5% of the global ocean). Furthermore, our
- estimate is only about half of that estimated by Wanninkhof et al. (2013), whose budget was based on extrapolating adjacent open-ocean data-based estimates of the specific flux into the coastal ocean. We attribute our lower specific flux of anthropogenic carbon into the global coastal ocean mainly to the model's associated offshore carbon transport, which is not strong enough to reduce surface levels of anthropogenic DIC (and thus anthropogenic *p*CO<sub>2</sub>) to levels that are as low as those in the
  open ocean (on average). Whether or not our model provides a realistic estimate of offshore transport at the global scale is a critical question, however, that demands further investigation.

Clearly, our approach is limited by the extent to which the coastal ocean is resolved. Our model's horizontal resolution does not allow it to fully resolve some fine-scale coastal processes such as tides, which affect  $FCO_2$  at tidal fronts (Bianchi et al., 2005). Model resolution is also inadequate to

30 fully resolve mesoscale and sub-mesoscale eddies and associated upwelling. Moreover, in the midlatitudes with a water depth of 80 m, the first baroclinic Rossby radius (the dominant scale affecting coastal processes) is around 200 km but it falls below 10 km on Arctic shelves (Holt et al., 2014; Nurser and Bacon, 2014). Thus the higher latitudes need much finer resolution (Holt et al., 2009). Yet all model studies must weigh the costs and benefits of pushing the limits toward improved realism. Our approach has been to use a model that takes only a first step into the eddying regime in order to be able to achieve long physical-biogeochemical simulations with atmospheric  $CO_2$  increasing from preindustrial levels to today. It represents a step forward when compared to studies

5 with typical coarse resolution ocean models (around 2° horizontal resolution), which may be considered to be designed exclusively for the open ocean. In the coming years, increasing computational resources will allow further increases in spatial resolution and a better representation of the coastal ocean in global ocean carbon cycle models.

Improvements will also be needed in terms of the modeled biogeochemistry of the coastal zone.

- 10 Most global-scale biogeochemical models neglect river input of nutrients and carbon. Although that is taken into account in our simulations, the river input forcing is constant in time (Aumont et al., 2015). Seasonal and higher frequency variability in carbon and nutrient river input (e.g., from floods and droughts) is substantial as often are anthropogenic trends. For simplicity, virtually all global-scale models neglect sediment resuspension and early diagenesis in the coastal-zone. Those processes in
- 15 some coastal areas may well alter nutrient availability, surface DIC, and total alkalinity, which would affect FCO<sub>2</sub>. In addition, in the coastal zone, one must eventually go beyond the classic definition of anthropogenic carbon, i.e., the change due only to the direct influence of the anthropogenic increase in atmospheric CO<sub>2</sub> on the FCO<sub>2</sub> and ocean carbonate chemistry. Changes in other human induced perturbations may be substantial. For example, an important research topic will be to better assess
  20 potential changes in sediment burial of carbon in the coastal zone during the industrial era, estimated
- at up to  $0.15 \text{ Pg} \text{ Cyr}^{-1}$  but with large uncertainty (Regnier et al., 2013).

To improve understanding of the critical land-ocean connection and its role in carbon and nutrient exchange, we call for a long-term effort to exploit the latest, global-scale, high-resolution, ocean general circulation models, adding ocean biogeochemistry, and improving them to better represent

25 the coastal and open oceans together as one seamless system.

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**Table 1.** Sea-to-air total  $CO_2$  fluxes ( $Pg C yr^{-1}$ ) given as zonal means from Takahashi et al. (2009) for the reference year 2000, from Landschützer et al. (2014) for 1998-2011 and from the ORCA05 model for 1993–2012.

Latitudinal	Observation-ba	This study		
bands	Takahashi et al. (2009)	Landschützer et al. (2014)	ORCA05	
90°S - 30°S	-0.77	-0.73	-1.50	
$30^{\circ}$ S - $30^{\circ}$ N	0.20	0.13	0.13	
$30^{\circ}$ N - $90^{\circ}$ N	-1.59	-1.53	-0.93	

**Table 2.** MARCATS regions as described by Laruelle et al. (2013, 2014), along with means for data-based fluxes of total CO<sub>2</sub> from LA14 during 1990-2011 as well as simulated anthropogenic and total CO<sub>2</sub> fluxes, and **residence time** during 1993–2012. **Uncertainties are the interannual variability over the averaged period.** Abbreviations are included for North (N), South (S), East (E), West (W), Eastern Boundary Current (EBC); Western Boundary Current (WBC), sea-to-air flux of total carbon (FCO<sub>2</sub><sup>tot</sup>), anthropogenic carbon (FCO<sub>2</sub><sup>ant</sup>). Surface areas indicated as 'from LA14' actually differ slightly from those published in LA13 as they have been modified for subsequent computations (Goulven G. Laruelle, personal communication, January 2015).

$\mathbf{N}^{\mathrm{o}}$	System Name	Class	Surface (	$10^{3}  \rm km^{2}$ )	FCO <sub>2</sub> <sup>tot</sup> (mol	$C m^{-2} yr^{-1}$ )	FCO <sub>2</sub> <sup>tot</sup> (Tg C	$yr^{-1}$ )	$FCO_2^a$	nt	Residence
			Model	LA14	Simulated	LA14	Simulated	LA14	$\rm molCm^{-2}yr^{-1}$	${\rm Tg}{\rm C}{\rm yr}^{-1}$	time (month)
1	N-E Pacific	Subpolar	397	350	$\textbf{-2.29}\pm0.17$	-1.61	$-10.935 \pm 0.823$	-6.775	$\textbf{-0.45} \pm 0.05$	$\textbf{-2.16} \pm 0.23$	$0.83\pm0.23$
2	Californian Current	EBC	118	208	$\textbf{-0.34} \pm 0.10$	-0.05	$\textbf{-0.477} \pm 0.148$	-0.135	$\textbf{-0.35}\pm0.09$	$\textbf{-0.50}\pm0.13$	$1.00\pm0.23$
3	Tropical E Pacific	Tropical	152	183	$\textbf{-0.12}\pm0.05$	0.09	$\textbf{-0.222}\pm0.095$	0.192	$\textbf{-0.36} \pm 0.05$	$\textbf{-0.65} \pm 0.10$	$0.51\pm0.09$
4	Peruvian Upwelling Current	EBC	138	143	$1.44\pm0.80$	0.65	$2.386 \pm 1.325$	1.073	$\textbf{-0.39}\pm0.09$	$\textbf{-0.64} \pm 0.15$	$0.72\pm0.15$
5	Southern America	Subpolar	1126	1190	$\textbf{-1.51}\pm0.13$	-1.31	$-20.460 \pm 1.705$	-18.715	$\textbf{-0.46} \pm 0.05$	$\textbf{-6.28} \pm 0.74$	$0.65\pm0.05$
6	Brazilian Current	WBC	475	484	$\textbf{-0.33} \pm 0.08$	0.10	$\textbf{-1.872}\pm0.479$	0.567	$\textbf{-0.34} \pm 0.05$	$\textbf{-1.95}\pm0.29$	$0.26\pm0.06$
7	Tropical W Atlantic	Tropical	479	488	$0.86\pm0.10$	0.07	$4.934\pm0.551$	0.394	$\textbf{-0.26} \pm 0.05$	$\textbf{-1.50}\pm0.31$	$0.20\pm0.02$
8	Caribbean Sea	Tropical	303	358	$0.10\pm0.10$	0.81	$0.366\pm0.348$	3.460	$\textbf{-0.31}\pm0.04$	$\textbf{-1.12}\pm0.14$	$0.32\pm0.03$
9	Gulf of Mexico	Marginal Sea	469	532	$\textbf{-0.79} \pm 0.11$	-0.33	$\textbf{-4.478} \pm \textbf{0.633}$	-2.100	$\textbf{-0.32}\pm0.03$	$\textbf{-1.81}\pm0.16$	$1.01\pm0.15$
10	Florida Upwelling	WBC	545	591	$\textbf{-2.25}\pm0.21$	-0.38	$-14.692 \pm 1.351$	-2.723	$\textbf{-0.66} \pm 0.05$	$\textbf{-4.29} \pm 0.36$	$0.39\pm0.02$
11	Sea of Labrador	Subpolar	576	638	$\textbf{-1.27}\pm0.18$	-1.72	$\textbf{-8.808} \pm \textbf{1.244}$	-13.172	$\textbf{-0.32}\pm0.03$	$\textbf{-2.19}\pm0.21$	$1.20\pm0.35$
12	Hudson Bay	Marginal Sea	998	1064	$0.31\pm0.29$	n.d	$3.757\pm3.423$	n.d.	$\textbf{-0.08} \pm 0.04$	$\textbf{-0.99}\pm0.46$	$51.22\pm22.75$
13	Canadian Archipelago	Polar	1001	1145	$\textbf{-0.52}\pm0.06$	-1.02	$\textbf{-6.234} \pm 0.748$	-13.986	$\textbf{-0.09}\pm0.02$	$\textbf{-1.03}\pm0.21$	$2.82\pm0.46$
14	N Greenland	Polar	544	602	$\textbf{-0.97} \pm 0.15$	-0.61	$\textbf{-6.333} \pm 1.000$	-4.400	$\textbf{-0.26} \pm 0.05$	$\textbf{-1.67}\pm0.33$	$2.38\pm0.44$
15	S Greenland	Polar	238	262	$\textbf{-3.35}\pm0.44$	-3.81	$\textbf{-9.564} \pm \textbf{1.259}$	-11.972	$\textbf{-0.86} \pm 0.19$	$-2.45\pm0.53$	$0.48\pm0.09$
16	Norwegian Basin	Polar	141	162	$\textbf{-2.87} \pm 0.23$	-1.72	$\textbf{-4.855} \pm 0.396$	-3.342	$\textbf{-0.60}\pm0.09$	$\textbf{-1.02}\pm0.15$	$0.31\pm0.10$
17	N-E Atlantic	Subpolar	1020	1073	$\textbf{-2.16} \pm 0.12$	-1.33	$-26.501 \pm 1.419$	-17.165	$\textbf{-0.53} \pm 0.05$	$\textbf{-6.52}\pm0.59$	$0.93\pm0.11$
18	Baltic Sea	Marginal Sea	324	364	$0.30\pm0.07$	0.51	$1.184\pm0.288$	2.245	$\textbf{-0.01} \pm 0.01$	$\textbf{-0.05}\pm0.03$	$17.37\pm9.52$
19	Iberian Upwelling	EBC	251	267	$-1.13\pm0.12$	0.04	$-3.393 \pm 0.352$	0.122	$-0.27\pm0.03$	$-0.82\pm0.10$	$2.31\pm0.54$
20	Mediterranean Sea	Marginal Sea	423	529	$\textbf{-0.24}\pm0.06$	0.62	$\textbf{-1.196} \pm 0.327$	3.925	$\textbf{-0.30}\pm0.02$	$\textbf{-1.52}\pm0.12$	$0.72\pm0.09$
21	Black Sea	Marginal Sea	131	172	$-0.24 \pm 0.11$	n.d.	$-0.375 \pm 0.174$	n.d.	$\textbf{-0.18} \pm 0.02$	$-0.28\pm0.03$	$1.60\pm0.48$
22	Moroccan Upwelling	EBC	177	206	$0.18\pm0.12$	2.92	$0.385\pm0.263$	7.220	$\textbf{-0.33}\pm0.03$	$\textbf{-0.71} \pm 0.07$	$0.67\pm0.14$
23	Tropical E Atlantic	Tropical	225	259	$0.09\pm0.08$	-0.06	$0.239 \pm 0.208$	-0.174	$\textbf{-0.19}\pm0.02$	$-0.52\pm0.05$	$0.59\pm0.09$
24	S W Africa	EBC	300	298	$0.43\pm0.40$	-1.43	$1.544 \pm 1.448$	-5.103	$\textbf{-0.59}\pm0.08$	$\textbf{-2.14}\pm0.28$	$2.17\pm0.55$
25	Agulhas Current	WBC	189	239	$\textbf{-1.20}\pm0.09$	-0.58	$\textbf{-2.730} \pm 0.206$	-1.664	$\textbf{-0.53} \pm 0.05$	$\textbf{-1.21}\pm0.12$	$0.13\pm0.01$
26	Tropical W Indian	Tropical	46	68	$\textbf{-0.06} \pm 0.08$	1.00	$\textbf{-0.031} \pm 0.044$	0.815	$\textbf{-0.16} \pm 0.04$	$\textbf{-0.09}\pm0.03$	$0.20\pm0.04$
27	W Arabian Sea	Indian Margins	82	92	$0.35\pm0.04$	1.14	$0.342\pm0.043$	1.257	$\textbf{-0.31} \pm 0.04$	$\textbf{-0.31}\pm0.04$	$0.12\pm0.04$
28	Red Sea	Marginal Sea	158	174	$0.24\pm0.03$	0.16	$0.460\pm0.065$	0.330	$-0.15\pm0.01$	$\textbf{-0.28} \pm 0.02$	$0.57\pm0.15$
29	Persian Gulf	Marginal Sea	208	233	$0.04\pm0.08$	n.d.	$0.092\pm0.203$	n.d.	$\textbf{-0.12}\pm0.02$	$\textbf{-0.31}\pm0.04$	$24.67 \pm 12.09$
30	E Arabian Sea	Indian Margins	298	317	$0.21\pm0.12$	0.67	$0.749 \pm 0.427$	2.555	$\textbf{-0.30}\pm0.04$	$\textbf{-1.07}\pm0.15$	$0.67\pm0.15$
31	Bay of Bengal	Indian Margins	197	203	$\textbf{-0.69} \pm 0.12$	-0.22	$\textbf{-1.641} \pm 0.276$	-0.530	$\textbf{-0.31}\pm0.04$	$\textbf{-0.74} \pm 0.09$	$0.43\pm0.11$
32	Tropical E Indian	Indian Margins	727	763	$\textbf{-0.06} \pm 0.07$	-0.02	$\textbf{-0.482} \pm 0.569$	-0.170	$\textbf{-0.20}\pm0.02$	$-1.78\pm0.17$	$0.50\pm0.04$
33	Leeuwin Current	EBC	81	117	$\textbf{-2.05}\pm0.15$	-0.98	$\textbf{-2.010} \pm 0.148$	-1.379	$-0.60\pm0.07$	$\textbf{-0.58} \pm 0.07$	$0.56\pm0.16$
34	S Australia	Subpolar	392	436	$\textbf{-1.37}\pm0.18$	-1.14	$\textbf{-6.438} \pm 0.859$	-5.983	$\textbf{-0.27}\pm0.03$	$\textbf{-1.29}\pm0.14$	$0.74\pm0.25$
35	E Australian Current	WBC	98	130	$\textbf{-1.74}\pm0.18$	-1.09	$-2.036\pm0.205$	-1.695	$\textbf{-0.50}\pm0.07$	$\textbf{-0.58} \pm 0.08$	$0.37\pm0.04$
36	New Zealand	Subpolar	263	286	$\textbf{-1.23}\pm0.16$	-1.25	$\textbf{-3.882} \pm 0.498$	-4.274	$\textbf{-0.52}\pm0.07$	$\textbf{-1.64} \pm 0.23$	$0.49\pm0.04$
37	N Australia	Tropical	2278	2292	$\textbf{-0.29}\pm0.11$	0.44	$-7.872 \pm 3.114$	12.120	$\textbf{-0.23}\pm0.04$	$\textbf{-6.19} \pm 1.00$	$0.38\pm0.03$
38	S-E Asia	Tropical	2130	2160	$\textbf{-0.29} \pm 0.07$	-0.91	$\textbf{-7.344} \pm 1.908$	-23.609	$\textbf{-0.20}\pm0.03$	$-5.01\pm0.72$	$0.49\pm0.05$
39	China Sea and Kuroshio	WBC	1132	1129	$\textbf{-1.99}\pm0.15$	-1.41	$-27.046 \pm 1.991$	-19.100	$\textbf{-0.45} \pm 0.05$	$\textbf{-6.13} \pm 0.72$	$0.32\pm0.01$
40	Sea of Japan	Marginal Sea	233	147	$-3.07\pm0.17$	-3.47	$\textbf{-8.613} \pm 0.475$	-6.113	$\textbf{-0.51}\pm0.06$	$-1.44\pm0.18$	$1.64\pm0.24$
41	Sea of Okhotsk	Marginal Sea	933	952	$\textbf{-1.66} \pm 0.07$	1.31	$-18.623 \pm 0.761$	14.955	$\textbf{-0.36} \pm 0.03$	$-4.00\pm0.34$	$3.52\pm 1.38$
42	N-W Pacific	Subpolar	1025	1000	$\textbf{-1.85}\pm0.14$	-0.70	$-22.760 \pm 1.726$	-8.419	$\textbf{-0.24} \pm 0.04$	$-2.99\pm0.52$	$1.48\pm0.59$
43	Siberian Shelves	Polar	1848	1889	$\textbf{-0.47} \pm 0.10$	-0.90	$-10.499 \pm 2.117$	-20.322	$\textbf{-0.05} \pm 0.01$	$\textbf{-1.09}\pm0.28$	$4.10\pm0.64$
44	Barents and Kara Seas	Polar	1559	1680	$\textbf{-0.75} \pm 0.14$	-1.60	$-14.176 \pm 2.585$	-32.225	$\textbf{-0.11}\pm0.02$	$-2.05\pm0.43$	$1.58\pm0.46$
45	Antarctic Shelves	Polar	2452	2936	$\textbf{-0.90} \pm 0.14$	-0.15	$-26.630 \pm 3.989$	-5.381	$\textbf{-0.69} \pm 0.07$	$\textbf{-20.30} \pm 2.18$	$2.08\pm0.29$

**Table 3.** Weighted mean of simulated and data-based sea-to-air CO<sub>2</sub> fluxes and **simulated residence time** for each MARCATS class, excluding the Sea of Okhotsk (see text). Abbreviations are included for Eastern Boundary Current (EBC) and Western Boundary Current (WBC).

Class	Sea-to	Residence		
	Total (LA14)	Total (model)	Anthropogenic (model)	time (month)
EBC	0.12	$\textbf{-0.12}\pm0.16$	$-0.42\pm0.03$	$1.52\pm0.22$
Indian margins	0.19	$\textbf{-0.06} \pm 0.05$	$\textbf{-0.24}\pm0.02$	$0.49\pm0.04$
Marginal Seas	-0.56	$\textbf{-0.92}\pm0.07$	$\textbf{-0.29}\pm0.01$	$10.34\pm3.50$
Polar Margins	-0.88	$\textbf{-0.83}\pm0.06$	$\textbf{-0.32}\pm0.03$	$2.18\pm0.20$
Subpolar Margins	-1.23	$-1.61\pm0.07$	$\textbf{-0.36} \pm 0.02$	$0.92\pm0.16$
Tropical Margins	-0.10	$\textbf{-0.15}\pm0.06$	$\textbf{-0.22}\pm0.02$	$0.42\pm0.03$
WBC	-0.80	$-1.65\pm0.08$	$-0.48\pm0.03$	$0.31\pm0.01$

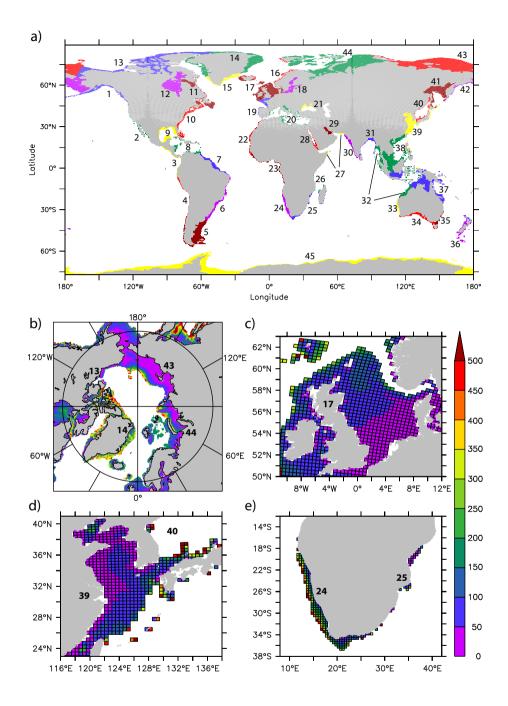
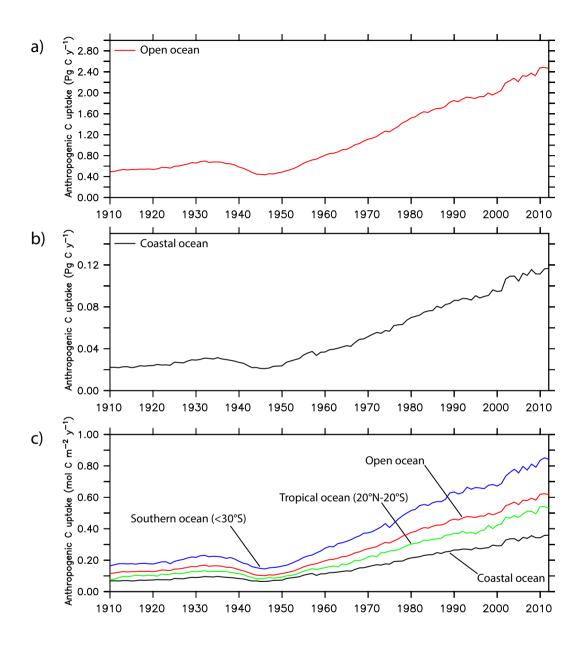
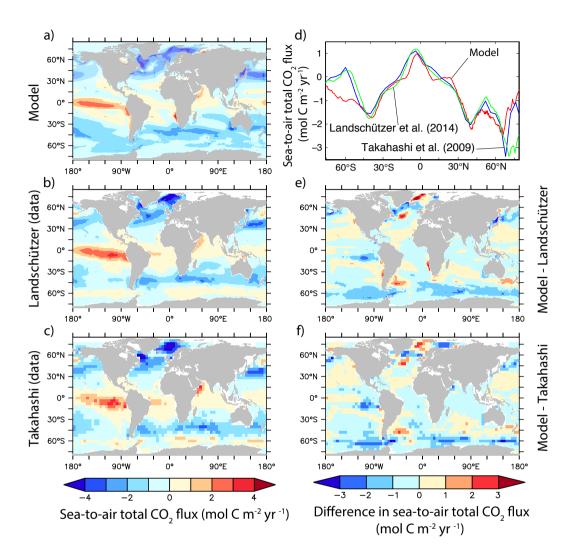


Figure 1. (a) Global segmentation of the coastal ocean following Laruelle et al. (2013) as regridded on the ORCA05 model grid. Colors distinguish limits between the MARCATS regions; numbers indicate regions defined in LA13. To perceive the spatial resolution of the ORCA05 configuration in the MARCATS context, we show zooms of bathymetry in 4 regions: (b) The Arctic polar margins, (c) the North Sea, (d) the Sea of Japan, the China Sea, and Kuroshio, and (e) Southern Western Africa and the Aghulas Current. In the latter 3 panels, grid resolution is indicated by thin black lines. 29



**Figure 2.** Simulated temporal evolution of area-integrated anthropogenic carbon uptake for (a) the **open** ocean and (b) the coastal ocean. (c) Analogous evolution of anthropogenic carbon uptake for the **open** ocean, the coastal ocean, the Southern Ocean, and the tropical oceans, but given as the average flux per unit area.



**Figure 3.** Climatological mean of sea-to-air flux of total carbon fluxes in mol C m<sup>-2</sup> y<sup>-1</sup> for (a) the model average during 1993–2012, (b) the data-based estimate from Landschützer et al. (2014) for 1998–2011, and (c) the data-based estimate from Takahashi et al. (2009) for the 2000–2009. Panels (d) and (f) present differences between simulated and observed sea-to-air total carbon fluxes (mol C m<sup>-2</sup> yr<sup>-1</sup>) relative to (b) and (c), respectively. d) presents the latitudinal distribution of the simulated and the observed mean sea-to-air total carbon fluxes.

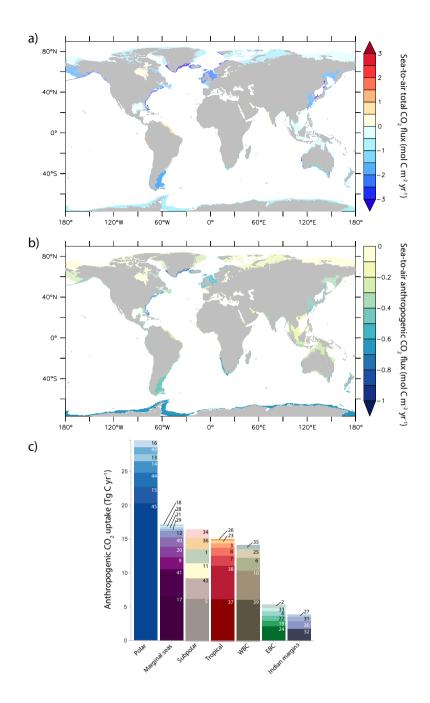
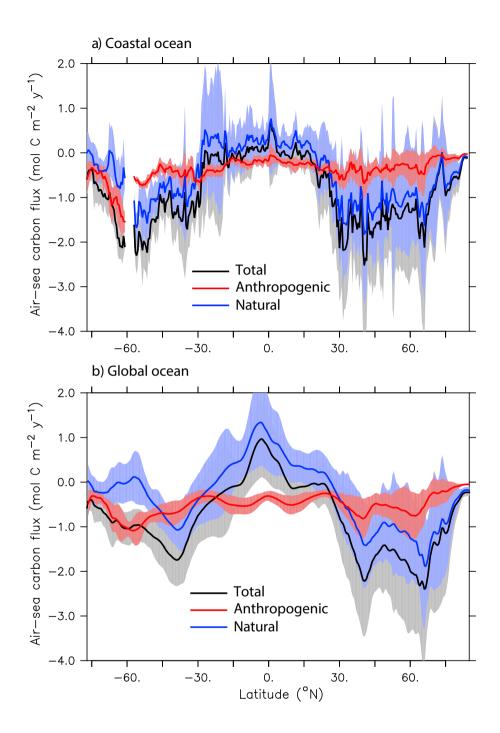


Figure 4. Global mean distribution of the simulated sea-to-air flux of (a) total carbon and (b) anthropogenic carbon over 1993–2012 as mol C m<sup>-2</sup> yr<sup>-1</sup> in the global coastal ocean segmented following MARCATS from LA13. (c) Bar chart of the anthropogenic carbon uptake in Tg C yr<sup>-1</sup> according to the MARCATS classification. Abbreviations are included for Eastern Boundary Current (EBC) and Western Boundary Current (WBC). Links between numbers and regions are reported in Table 2. Interactive illustrations can be found at http://lsce-datavisgroup.github.io/CoastalCO2Flux/.



**Figure 5.** Zonal-mean, sea-to-air fluxes of total, anthropogenic, and natural  $CO_2 \pmod{cm^{-2} yr^{-1}}$  given as the average over 1993–2012 for (a) the coastal ocean and (b) the global ocean. Shaded areas indicate the standard deviation of environmental variability of all ocean grid cells within each latitudinal band. Interannual variations are not shown.

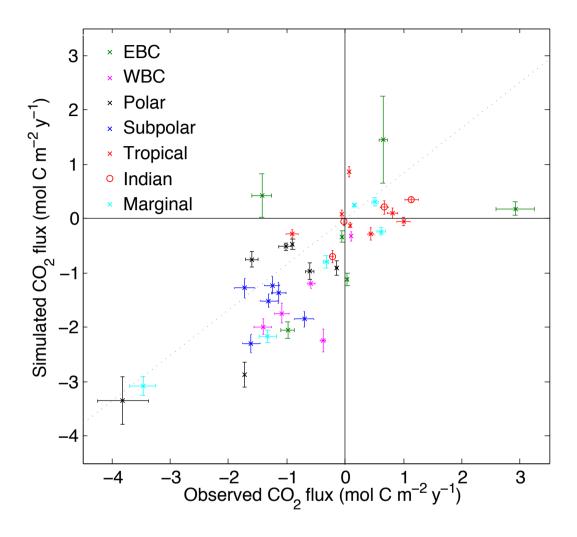
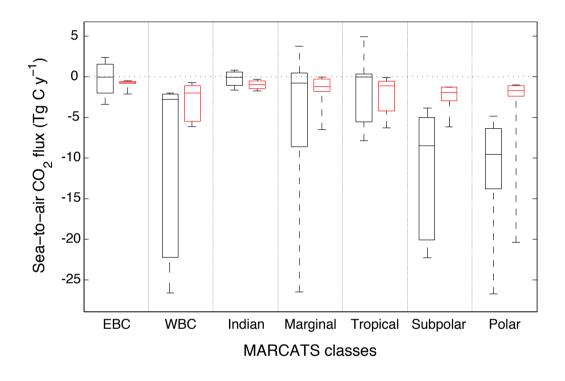


Figure 6. Simulated versus observed MARCATS sea-to-air flux of total carbon in (a)  $mol C m^{-2} yr^{-1}$  and (b)  $Tg C yr^{-1}$ . Vertical error bars show the standard deviation from the 1993–2012 interannual variability for model results and the horizontal bars correspond to the 1990–2011 variability from computational methods used in LA14 for observation-based estimates. Here, regression line (grey dotted) have *y*-intercepts forced to 0. All MARCATS regions have been used except the Black Sea, the Persian Gulf (no data estimate), and the Sea of Okhotsk (see text); corrections were also applied for the Florida-Labrador delimitation.



**Figure 7.** Box plots of the simulated sea-to-air  $CO_2$  fluxes  $(Tg C yr^{-1})$  grouped into the MARCATS classes of the coastal ocean. Black boxes indicate total fluxes; red boxes indicate anthropogenic fluxes. Shown are the lowest estimate, the first quartile, the median, the third quartile, and the highest estimate for each class.

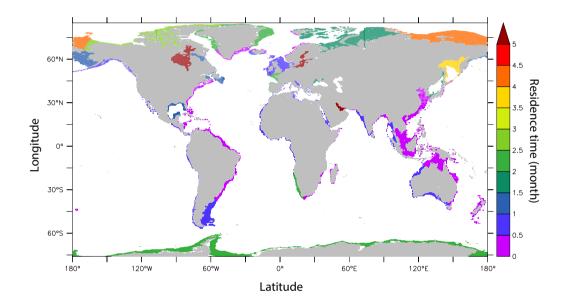
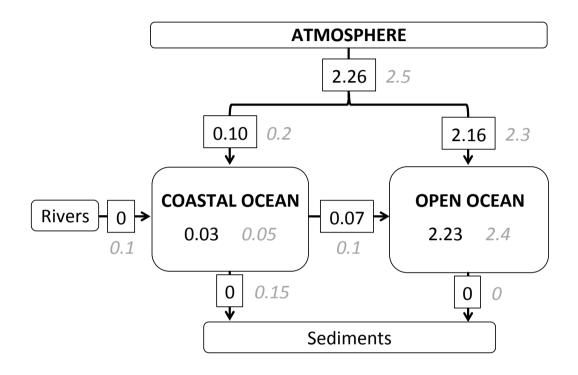


Figure 8. Global distribution of simulated residence time (month) for the global coastal ocean segmented following Laruelle et al. (2013).



**Figure 9.** Transfer of anthropogenic carbon between the atmosphere, coastal ocean, and open ocean along with increases in the corresponding inventory in each reservoir, given as the average of simulated values over 1993–2012. All results are in  $Pg C yr^{-1}$ . Simulated results are shown as dark numbers in boxes; adjacent numbers (grey italic) indicate data-based estimates for the 2000–2010 average (Regnier et al., 2013).

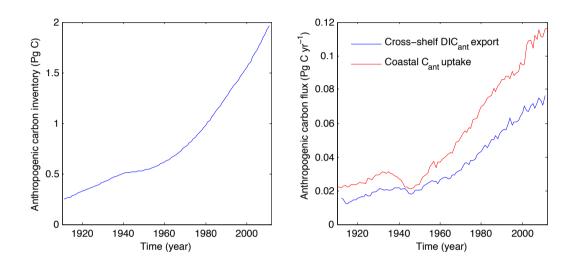


Figure 10. Simulated temporal evolution of (a) coastal-ocean inventory of anthropogenic carbon given in Pg C and (b) anthropogenic  $CO_2(C_{ant})$  uptake by the global coastal ocean and global cross-shelf export of anthropogenic carbon ( $DIC_{ant}$ ) given in Pg C yr<sup>-1</sup>.