Answer to Reviewer #1

After reviewing this manuscript, I do not know that weather I should believe the estimated results for the mean annual net ecosystem metabolism (NEM), FCO2, and CFilt or not, because it is a big issue for air-sea interaction. I am confusing that the authors tried to use a simple 1D model coupled with Global News model in current study. How did they do? Many assumptions should be made to compromise the estimated results for air-sea CO2 evasion. The authors should clarify many assumptions in their study. In the model, many parameters should be set up to simulate the state variables shown in Table 2. How did the authors select those parameters? The parameters and values should be listed clearly.

We agree with the reviewer that a better representation of carbon dynamics through the quantification of the Net Ecosystem Metabolism, CO_2 outgassing and carbon filtration in estuarine systems is a critical issue for air-sea interaction. This topic is a particularly pressing matter at the regional scale due to the difficulty of deriving consistent regional budgets from the upscaling of rare local measurements performed in morphologically complex and profoundly heterogeneous systems (Borges and Abril, 2011; Laruelle et al., 2013; Regnier et al., 2013). On the modeling side, the set-up of a reliable reactive transport model able to realistically capture the estuarine carbon dynamics generally proves a very costly endeavor in terms of data requirement to constrain the model (i.e. bathymetric data, boundary conditions, climatic forcing…) and in terms of time necessary to develop such model and run it (see e.g. Garnier et al., 2001; Huret et al., 2005; Arndt et al., 2011; Mateus et al., 2012). The model presented here is thus developed as a compromise, as it is currently the only one capable of running regional scale simulations with limited data and computation needs without sacrificing too much to oversimplification (as done when using box models to represent estuarine systems, Gordon et al., 1996). It follows from several studies published over the past few years (Regnier et al., 2013; Volta et al., 2014, 2016a, 2016b) that led to the development of a 1 dimensional generic estuarine model for tidal systems (Volta et al., 2014) forced by a set of generic parameters compiled from an unprecedented literature review (Volta et al., 2016a). This model was successfully applied and validated on several European estuaries (the Scheldt and Elbe, in particular, see Volta et al., 2016a&b) as well as at the regional scale of the North Sea, using a strategy similar to that presented here (Volta et al., 2016b). This strategy involved the use of the same boundary conditions as those used here for the east coast of the US. That is, the outputs of the global river model GLOBALNEWS and the global river carbon database GloRiCH to constrain upstream boundary conditions and the use of the World Ocean Atlas to specify the downstream boundary conditions. In other words, the model described in our

manuscript has precisely been designed to produce regional estuarine carbon budgets using the outputs of GlobalNEWS as boundary conditions and was already successfully used for similar purpose in another region.

As a consequence of the reviewer's skepticism and following numerous precise suggestions form the other reviewer, we have substantially modified the manuscript to better describe and justify our methodology, its underlying assumptions and potential limitations. We have also made the set-up of our simulations more transparent to secure reproducibility of our model results. In particular, the updated version of the manuscript now contains:

- A substantially modified introduction that puts our study into a more precise context and provides an improved description of the structure of the manuscript.

- Numerous additions to the model description section in order to clarify and substantiate all the assumptions on which our model relies on (i.e. calculation of boundary conditions, period of simulation, choice of databases, etc…), and which together, describe in much more detail the set-up of our simulations.

- 6 comprehensive tables (presented in the supplementary information) and which contain all physical forcings (i.e. estuarine geometry, wind speed, temperature…) and boundary conditions (nutrients and carbon concentrations, pH, alkalinity…).

- A new section (3.3. Scope of applicability and model limitations) which reflects on the strength and weaknesses of our modeling strategy in light of the current state of knowledge available to constrain a model such as ours. In particular, the adequacy of our approach to tackle regional scale modeling, the set-up of boundary condition with available databases and the quantification of the model's uncertainty are addressed in this section.

In 2.6 Model-data comparison, the description of this subsection is very poor. The authors described the model validation for other estuaries in the Europe. How did the authors validate the model for the study areas (U.S. east coast estuaries)? I would like to see the model validation in the study areas to convince me the model is capable and suitable to be used in U.S. east coast estuaries.

We understand the reviewer's concern about the limited validation of our model within the study area. This issue was also pointed out by reviewer #2. We thus expanded extensively section 2.6 to confront the annual CO_2 outgassing predicted by our model with 13 published estimates derived from direct measurements performed in estuaries located along the East coast of the US (Table 1). In addition, we provide a validation of our hydrodynamic model using several seasonal longitudinal salinity profiles in the Delaware Bay as well a validation of our biogeochemical model for two estuaries (the Delaware Bay and the Altamaha estuary). These additional simulations

reveal that C-GEM is able to properly represent a pCO_2 (Delaware Bay) and both pH and pCO_2 longitudinal profiles along the estuarine gradient (Altamaha). Also, in the new section 3.3 (Scope of applicability and model limitations), a paragraph discusses the issue of representativeness of the model's performance through local punctual validations in the case of regional simulations including numerous small systems for which the data that would be required to perform a local validation are simply inexistent.

We hope that all these modifications will convince the reviewer of the usefulness and relevance of our study and modelling strategy.

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Volta, C., Laruelle, G. G., and Regnier, P.: Regional carbon and CO2 budgets of North Sea tidal estuaries, Estuarine, Coastal and Shelf Science, 176, 76-90, 2016a.

Volta, C., Laruelle, G. G., Arndt, S., and Regnier, P.: Linking biogeochemistry to hydro-geometrical variability in tidal estuaries: a generic modeling approach, Hydrol. Earth Syst. Sci., 20, 991-1025, doi:10.5194/hess-20-991-2016, 2016b.

Answer to Reviewer #2

Overall statements

The manuscript "Air-water CO2 evasion from U.S. East Coast estuaries" by Goossens, N., Gildas, L.G., Arndt, S., Regnier, P. gives valuable estimates on the main biogeochemical fluxes of the estuaries along the US east coast. The authors model 43 tidal estuaries and subdivide the results into 3 different latitudinal zones showing distinct differences which appear reasonable. The problem is that the reader has to accept these "black box results" even though all the details of the different estuaries should be available. I will pinpoint the problems and possible ways to resolve them:

We are grateful for the reviewer's evaluation and the constructive suggestions provided. We understand that the reviewer is mainly concerned about an apparent 'lack of transparency', as well as a seemingly weak validation of the model within the study area. Following the reviewer's recommendations, we thus substantially modified the manuscript to respond to these concerns. More specifically, we added a comparison between model-predicted annual CO_2 outgassing fluxes and 13 published flux estimates, derived from direct measurements in local estuaries to section 2.6 (Model-data comparison). In addition, we also provide new validations of the hydrodynamic and biogeochemical model (section 2.6). Furthermore, we introduced a new section (section 3.4), which critically discusses the scope of applicability and model limitations.

Please find bellow a detailed answer to each comment. All our answers are written in blue and the modifications within the text are highlighted in bold and italic. In the revised manuscript, changes are tracked via Word's track changes tool.

On behalf of all co-authors,

Goulven Laruelle

* The data preparation for the 43 estuaries is not transparent and reproducible. Please prepare a table in which all details for each estuary are inserted (like Volta et al., 2016a, Tab. 1). If this table appears too large, put it into the Appendix (supplemental data).

In the revised manuscript, we now provide 5 additional, extensive tables as supplementary information, which summarize all key parameters and boundary conditions required to perform the simulations. In addition to table SI1, which already provided the estuarine surface areas, as well as fresh water discharge fluxes for all systems and seasons, these new tables provide:

Table SI2: Geometric properties of the estuary (i.e. length, width at both boundaries, depth and convergence length)

Table SI3: Upstream boundary conditions for nutrients and chlorophyll concentrations

Table SI4: Downstream boundary conditions for nutrients and chlorophyll concentrations

Table SI5: Upstream boundary conditions for the organic carbon and carbonate system (i.e. TOC, DIC, pH…)

Table SI6: Downstream boundary conditions for the organic carbon and carbonate system (i.e. TOC, DIC, pH…)

Please use lat/lon positions of the mouth and estuary names if possible.

As requested, latitudes and longitudes, as well as the names of the largest rivers are provided for each estuary in all aforementioned tables. In addition, within the main text, we now make reference, whenever possible, to the name and coordinates of the estuaries that are being discussed.

* The validation chapter only refers to applications elsewhere. Please validate the model for at least one estuary in each latitudinal zone like Volta et al., 2016a did it for some North Sea estuaries.

The general performance of C-GEM in reproducing and predicting estuarine hydrodynamics and biogeochemical cycling has been extensively tested across a large range of different estuarine systems (e.g. Volta et al., 2014, 2016, see also Savenije 2001 for the estuarine physics). Here, we extended these tests by a number of local model-data comparisons. We added a new comparison between model-predicted annual CO_2 outgassing fluxes and 13 published flux estimates, derived from direct local measurements to section 2.6 (Table 1). In addition, we also evaluated the performance of the hydrodynamic model by comparing simulation results with seasonal, longitudinal salinity profiles in the Delaware Bay. Furthermore, the performance of the biogeochemical model is critically evaluated by comparing simulation results with longitudinal profiles of pCO_2 and pH, in the Delaware Bay and the Altamaha River estuary. These additional model-data comparisons reveal that C-GEM is able to reproduce local measurements of pCO_2 (Delaware Bay), as well as longitudinal pH and pCO_2 profiles (Altamaha). Following what was done in Volta et al. (2016) with the Scheldt and the Elbe in Europe, the choice of using the Delaware Bay and the Altamaha river estuary was motivated by their contrasting geometries: The Delaware Bay is a marine dominated system characterized by a pronounced funnel shape while the Altamaha River ends with a very prismatic estuary characteristic of river dominated systems (Jiang et al., 2008). Thus, selecting these two end-members estuaries reveals the ability of C-GEM to simulate widely differing estuarine dynamics. Although we agree that performing a simulation on a system located in the Northern region would be a valuable addition, we could not find suitable a suitable set of observed nutrient and carbon boundary conditions and their corresponding longitudinal profiles. Note, however, that several flux values reported in Table 1 refer to estuaries located in this region. Finally, within the new section 3.4 (Scope of applicability and model limitations), we critically discuss the difficulties associated with 'validating' regional/global model simulations with a limited set of local, instantaneous observations, as well as the uncertainties that arise from the proposed model approach.

See updated manuscript at the end of this document

* You used some arguable boundary conditions and forcing functions: The Alkalinity near the mouth, DIC and Alkalinity from GLORICH positions closest to the river boundary or older discharge estimates. I know that it is difficult to put this all together in a reasonable way. But the reader should get knowledge about the sensitivities of the model in relation to estimates of boundary or forcing data. Please show how the model reacts on changes in these data. In the detailed statements I will show in which context such studies should be done.

As pointed out by the reviewer, designing realistic and consistent boundary conditions is a critical step in model set-up. In the present study, this task is further complicated by the necessity to find complete sets of boundary conditions for 43 tidal systems located along the eastern coast of the US. C-GEM was specifically designed with this difficulty in mind. Some of the strengths of the model are its comparatively modest data requirement and its transferability from one estuarine system to another, which has already been demonstrated on the estuaries surrounding the North Sea (Volta et al., 2016). C-GEM is also well suited to be operated in conjunction with global databases such as GlobalNEWS (Mayorga et al., 2010) because they share in common the watershed as their fundamental unit , which is essentially what was done in this study. We agree with the reviewer that some of the assumptions and choices of data sources could be better justified and critically discussed in the manuscript. We thus carefully addressed all concerns raised in the answers to the detailed statements of the reviewer. In addition, we also added an entire new section to the manuscript ('Scope of applicability and model limitations') that summarizes the strengths and weaknesses of our regional model approach.

As different input parameters are means over several years, the time span of validity of the results should be defined.

In the present study, simulations are representative for the year 2000 because some the largest datasets we rely on to constrain boundary conditions or forcings (e.g. GlobalNEWS) are derived from models calibrated for that year. As a consequence, additional data used to constrain boundary conditions and forcing parameters was selected from the same time span. In the revised manuscript, we added a few sentences justifying our data choice and specifying the time period for which simulation results are representative. In addition, we also specify if boundary conditions/forcings are constrained on the basis of punctual measurements or averages of several years.

The introduction reads rather as an advertising text. Give, for example, details about the structure of the ms. A question, which could be tackled, is whether global models miss estuarine processes (Line 39).

In the previous version of our introduction, we tried to emphasize the originality and the potential of our modelling approach. We felt that it was important to stress the novelty of the approach developed here: an explicit simulation of seasonal carbon transformations and fluxes along the land-ocean continuum at a regional scale. This work builds on the study of Volta et al. (2016) which provided the first annually average estimates of estuarine carbon transformations and fluxes focusing on the estuaries surrounding the North Sea. However, we agree with the reviewer that the proportion of the introduction dedicated to the presentation of C-GEM was too long and sometime superfluous. Following the reviewer's advice, we shortened the introduction and emphasized the research questions that can be tackled with the presented model approach. We also provided a better overview of the structure of the manuscript and the studied area. Furthermore, we followed the reviewer's suggestion and shortly discuss the ability of global carbon cycle models to account for the influence of the estuarine modulator.

See updated manuscript at the end of this document

Detailed statements

L14/15 Write 697.000 km2

Done

L19 For which time period?

The sentence has been re-written to state that model simulations are representative of the year 2000.

"***Our simulations, performed using conditions representative of the year 2000, suggest that,*** together, US East coast estuaries emit 1.9 TgC yr^{-1} ***in the form of CO_2***, which correspond to about 40 % of the carbon inputs from rivers, marshes and mangrove"

L19 Only CO2, or also other gases including carbon?

Model simulations only account for CO_2 exchange at the air-water interface and the sentence has been modified to clarify this point.

"…, together, US East coast estuaries emit 1.9 TgC yr^{-1} ***in the form of CO_2***, which correspond to about 40 % of the carbon inputs from rivers, marshes and mangroves."

L25 the results

Done

"Finally, ***the*** results reveal that the ratio of estuarine surface area to the river discharge, S/Q…"

L100 Make a full sentence: For a review see Laruelle et al. (2013)

We added a sentence.

"These comprehensive data sets are complemented by local observations of carbon cycling and CO_2 fluxes in selected, individual estuarine systems, making the East coast of the United States an ideal region for a first, fully explicit regional evaluation of CO_2 evasion resolving every major tidal estuary along the selected coastal segment. ***An extensive review of published local estimates of CO_2 fluxes in estuarine systems worldwide can be found in Laruelle et al. (2013).***"

L107/109/124 unify "Fig. x" -> all over the text

On the website of Biogeosciences, the guidelines for authors states: 'The abbreviation "Fig." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence, e.g.: "The results are depicted in Fig. 5. Figure 9 reveals that...".'

In the updated manuscript, we paid attention to strictly follow these rules.

L125 give lat/lon for these stations or enlarge Fig. 1 and indicate individual stations.

In the paragraph summarizing the published annual mean FCO_2 estimates based on measurements for Atlantic US estuaries, we replaced the reference to Figure 1 by a reference to Table 3, which provides a list of all the estuaries, as well as their respective coordinates mentioned in that section. Table 3 thus becomes Table 2 in the manuscript.

"A total of thirteen local, annual mean estuarine CO_2 flux estimates across the air-water interface based on measurements are also reported in the literature and are grouped along a latitudinal gradient ***(Tab. 2)***. "

L146ff 47 stationed were simulated. This contradicts the number of 43 (abstract).

Some watersheds flow into the same estuarine system and were merged to calculate boundary conditions for some of the systems (see section 2.4). The sentence the reviewer refers to mistakenly refers to the number of watersheds represented in our simulations rather than the number of estuaries actually represented. Only 43 tidal estuarine systems were simulated and the text was updated in sections 2.2, 2.3 and 3.4. The abstract, which refers to 43 estuaries, is thus still correct. We made the following corrections in the manuscript:

Line 147:

"The National Estuarine Eutrophication Assessment (NEAA) survey (Bricker et al., 2007), which uses geospatial data from the National Oceanic and Atmospheric Administration (NOAA) Coastal Assessment Framework (CAF) (NOAA, 1985), was used to identify and characterize ***58*** estuarine systems discharging along the Atlantic coast of the United States. From this set, ***43*** 'tidal' estuaries, defined as a river stretch of water that is tidally influenced (Dürr et al., 2011), were retained (fig.1) to be simulated by the C-GEM model, which is designed to represent such systems."

Line 171:

"The generic 1D Reactive-Transport Model (RTM) C-GEM (Volta et al., 2014) is used to quantify the estuarine carbon cycling in the ***43*** systems considered in this study."

Line 278:

"First, ***43*** coastal cells corresponding to tidal estuaries are identified in the studied area (Fig. 1)."

Line 568:

"The overall carbon filtering capacity of the region thus equals 41% of the total carbon entering the ***43*** estuarine systems (river + saltmarshes)."

15+47 is not 64 as I would expect from this sentence.
This is indeed a mistake, only 58 estuarine systems are presented (15+43, see comment above).

L151 Do you have a reference for this?

This figure was calculated using the GLOBALNEWS data (Mayorga et al., 2010) for POC and DOC combined with data from Hartmann et al. (2009) for DIC. We modified the sentence in a way that reflects the fact that we performed this calculation ourselves.

"***Using outputs from terrestrial models (Hartmann et al., 2009; Mayorga et al., 2010), the cumulated riverine carbon loads for all the*** non-tidal estuaries that are excluded from the present study ***amount to 0.9 Tg C yr^{-1} , which represents less than 15% of the total riverine carbon loads of the region. These 15 systems*** are located in the SAR (10) and in the MAR (5)."

L152 Tab. x not table x (all over the text)

We update the entire manuscript.

L169 2.9 m (use space)

Done

L207 "These parameters were determined through.."?

The geometric parameters we are referring to can be extracted using Geographic Information Systems (GIS). This widely used type of software allows the determination of, for example, a distance such as the width of an estuary at its mouth from a digitalized map. The manuscript has been modified to spell out GIS on its first occurrence and make clear that GIS is the tool that can be used to extract the data from local maps.

"These parameters can be easily determined ***from local maps or Google Earth using Geographic Information Systems (GIS)*** or obtained from databases (NASA/NGA, 2003)".

L210 are described

Corrected

L226 Use C only for concentration

C is the commonly used symbol for Chézy's coefficient but, to avoid any confusion with concentration, we followed the reviewer's advice and use Cz to denote the Chézy's coefficient. We updated equations (6) and the text.

L227 You mean eqs (5) and (6)?

Correct. The text was modified accordingly

L233 You mean eqs (5) and (6)?

Correct. The text was modified accordingly

L239 Use only English peer reviewed references

The original Dutch reference (Van der Burgh, 1972) was replaced by Savenije (1986), which is the oldest English peer reviewed publication using Van der Burgh's equations to calculate the dispersion coefficient in estuarine systems.

"The effective dispersion at the estuarine mouth can be quantified by the following relation ***(Savenije, 1986)***:"

L241 Define N by an equation

As stated in the text, the Canter Cremers' estuary number N corresponds to the ratio of the freshwater entering the estuary during a tidal cycle to the volume of salt water entering the estuary over a tidal cycle. We introduced a new equation (new equation 9) to define this parameter.

"…where h0 (m) is the tidally-averaged water depth at the estuarine mouth and N is the dimensionless Canter Cremers' estuary number defined as the ratio of the freshwater entering the estuary during a tidal cycle to the volume of salt water entering the estuary over a tidal cycle ***(Simmons, 1955)***:

$$N = \frac{Q_b \cdot T}{P}$$

In this equation, Q_b is the bankfull discharge ($m^3\ s^{-1}$), T is the tidal period (s) and P is the tidal prism (m^3). For each estuary, N can thus be calculated directly from the hydrodynamic model. "

Reference

Simmons, H. B.: Some effects of inland discharge on estuarine hydraulics, Proc. Am. Soc. Civ. Eng.-ASCE, 81, 792, 1955.

L260 Omit brackets

Done

L262 2000 m (use space)

Done

L272 273 please give a more detailed description here

The calculation of the gross primary production in the water column relies on a depth integration of the Platt equation (Platt et al., 1980), which assumes an exponential decrease of the light availability with depth. The article we are referring to (Vanderborght et al., 2007) described a cost efficient algebraic method to perform this integration using a gamma function. A sentence has been added to the text to provide more information about this procedure.

"The primary production dynamics, which requires vertical resolution of the photic depth, is calculated according to the method described in Vanderborght et al. (2007). ***This method assumes an exponential decrease of the light in the water column (Platt et al., 1980), which is solved using a Gamma function.***"

L276 For which year? Or are climatological or mean values used?

Boundary conditions for regional simulations that cover a large number of individual and sometimes underexplored systems are notoriously difficult to constrain. Comprehensive, temporally resolved observational data sets that would allow informing boundary conditions are rarely available. Global databases, such as GlobalNEWS, which provide model derived river loads of carbon and nutrients (Seitzinger et al., 2005; Mayorga et al., 2010) are a suitable alternative when direct observations are not available. Here, we used the GlobalNews database, the GloRich database and the World Ocean Atlas to constrain boundary conditions. GlobalNews model simulations have been calibrated for the year 2000. Boundary conditions for Alkalinity and pH are constrained on the basis of data extracted from the GloRiCh database. Most of the data compiled in this data base has been collected between 1990 and 2010. Finally, marine boundary conditions and water temperatures are derived the World Ocean Atlas. These values also correspond to climatological means centred on the year 2000. We explicitly paid attention to extracting coherent data sets from the respective databases that are representative of the same time period (around year 2000). We now clearly state in the main text which time period our boundary conditions are representative of:

"Boundary and forcing conditions are extracted from global databases and global model outputs that are available at 0.5° resolution. Therefore, C-GEM simulations are performed at the same resolution according to the following procedure. First, ***43*** coastal cells corresponding to tidal estuaries are identified in the studied area (***Fig.*** 1). If the mouth of an estuary is spread over several 0.5° grid cells, those cells are regrouped in order to represent a single estuary (e.g. Delaware estuary), and subsequently, a single idealized geometry is defined as described above. ***The model outputs (Hartmann et al. , 2009; Mayorga et al., 2010) and databases (Antonov et al., 2010; Garcia et al., 2010a; Garcia et al., 2010b) used to constrain our boundary conditions are representative of the year 2000.***"

L289 cloud coverage: Which is the origin of this data?

The data comes from ISCCP Cloud Data Products (Rossow and Schiffer, 1999) and the text was updated in order to include this reference:

"Mean daily solar radiation and photoperiods (corrected for cloud coverage ***using the ISCCP Cloud Data Products, Rossow and Schiffer, 1999***) are calculated depending on latitude and day of the year using a simple model (Brock, 1981).

Reference:

Rossow, W.B., and Schiffer, R.A., 1999: Advances in Understanding Clouds from ISCCP. Bull. Amer. Meteor. Soc., 80, 2261-2288."

L293 Are there no recent data available?

The UNH/GRDC runoff dataset of Fekete et al. (2002) is the dataset used in the GlobalNEWS simulations (Mayorga et al., 2010). Although these values are derived from long term averages over the 1960-1990 period, they have been adjusted to represent the year 2000 in the GlobalNEWS simulations. Here, we use the adjusted values to ensure that boundary conditions are representative of the same time period. In addition, while more recent databases exists such as the National Water Information System (NWIS), they do not provide water discharge for all US rivers and, thus, do not provide values for the totality of the systems used in our simulations.

L320 You mean 50 g C (g Chla)-1 ?

Correct. We meant 50 grams of carbon per gram of chlorophyll a and we updated the text with the notation suggested by the reviewer.

"The same source is used for phytoplankton concentrations, using a chlorophyll-a to phytoplankton carbon ratio of ***50 gC (gChla)$^{-1}$*** (Riemann et al., 1989) to convert the EPA values to carbon units used in the present study."

L332 339 W is not consistently defined. Is it percentage or surface area?

W corresponds to the fraction of the estuarine surface area represented by wetlands. The sentence introducing equation (11) has been updated to clarify this point:

"The DOC input of estuarine wetlands (Fig. 5b) scales to ***their fraction, W, of the total estuarine*** surface area and is calculated using the GlobalNEWS parameterization"

L341 give definition of "a"

The parameter 'a' is a calibration parameter used in the GlobalNEWS models and was first defined in Harrison et al. (2005) as: "unitless coefficient defining how non-point DOC export responds to runoff; for NEWS-DOC 'a' was set equal to 0.95."

In our calculations, we kept the same parametrization and rephrased the sentence as follows:

"W is the percentage of the land area within a watershed that is covered by wetlands, R is the runoff (m y-1) and a is a unit-less ***calibration*** coefficient defining how non-point source DOC export responds to runoff. ***The value of a is set to 0.95, consistent with the original GlobalNEWS -DOC model of Harrison et al. (2005)***."

L355 358 It seem that you use this argument twice. Here a sensitivity analysis would help.

The sentences pointed out by reviewer states that our models ignores DIC fluxes from tidal marshes because all the carbon exports from those systems come under the form of organic carbon and then justifies this assumption with the fact that very little degradation of organic matter takes place in the tidal marshes before it reaches the estuary. Considering the relatively small amounts of carbon that can be degraded within tidal marshes (and the lack of data to constrain such process) because of the short time scale of mixing processes, our assumption implicitly means that some biogeochemical processing taking place within tidal marshes may be accounted for by our estuarine model.

L364 I doubt that zero concentration for org C is appropriate at open sea boundary. Often org C is transported from the open sea into the estuaries were it is degradated.

Please substantiate this assumption.

We agree that the assumption that the open ocean is devoid of organic carbon is simplistic and that some organic carbon can enter the estuarine system from the seaward boundary. However, the focus of our study is to investigate and quantify the fate of the carbon delivered to estuaries by the riverine network. We introduced a sentence to section 2.4.4 to reflect on this limitation.

"This approach also reduces the influence of marine boundary conditions on the simulated estuarine dynamics, especially for all the organic carbon species whose concentrations are fixed at zero at the marine boundary. ***This assumption ignores the intrusion of marine organic carbon into the estuary during the tidal cycle but allows focusing on the fate of terrigenous material and its transit through the estuarine filter.***"

In addition, the implications of this assumption are also discussed in a paragraph of the new section 3.4:

"***C-GEM places the lower boundary condition 20 km from the estuarine mouth into the coastal ocean and the influence of this boundary condition on simulated biogeochemical dynamics is thus limited. At the lower boundary condition, direct observations for nutrients and oxygen are extracted from databases such as the World Ocean Atlas (Antonov et al., 2014). However, lower boundary conditions for OC and pCO_2 (zero concentration for OC and assumption of pCO_2 equilibrium at the sea side) are simplified. This approach does not allow addressing the additional complexity introduced by biogeochemical dynamics in the estuarine plume (see Arndt et al., 2011). Yet, these dynamics only play a secondary role in the presented study that focuses on the role of the estuarine transition zone in processing terrestrial-derived carbon."***

L374 domain -> boundary

Done

L377 Why longitudinal profiles? You mean at right angles with the river flow?

The term longitudinal profile is commonly used and refers to a concentration profile along the longitudinal (length) axis of the estuary, i.e. from the estuarine mouth to the river. Concentrations are thus representative of the cross-sectional average at the respective longitudinal position.

L383 How large do you estimate the error when neglecting degradation or burial in bottom sediments? A sensitivity test could help.

We agree that neglecting benthic processes is a potential limitation of our model. As the reviewer points out, organic matter degradation and burial may influence the biogeochemical of carbon in some estuaries and affect carbon retention within the system. However, because of the dynamic nature of estuarine sediments and the logistic challenges involved to sample them, direct observations and measurements of benthic processes are even more limited than those available for pelagic processes. Very little is known on the long term fate of organic carbon in estuarine sediments and its burial. Because of this lack of knowledge, benthic processes are not explicitly represented in the model. However, to a certain degree model parameters (such as organic matter degradation, denitrification rate constant) implicitly account for benthic dynamics. We acknowledge that, by ignoring benthic processes and burial in particular, our estimates for the estuarine carbon filtering may be underestimated. These considerations have been incorporated into a paragraph of the new section 3.4:

"***Although the reaction network of C-GEM accounts for all processes that control estuarine FCO_2 (Borges and Abril, 2012; Cai, 2011), several, potentially important processes, such as benthic-pelagic exchange processes, phosphorous sorption/desorption and mineral precipitation, a more complex representation of the local phytoplankton community, grazing by higher trophic levels, or multiple reactive organic carbon pools are not included. Although these processes are difficult to constrain and their importance for FCO_2 is uncertain, the lack of their explicit representations induces uncertainties in Cfilt. In particular, the exclusion of benthic processes such as organic matter degradation and burial in estuarine sediments could result in an underestimation of Cfilt. However, because very little is known on the long term fate of organic carbon in estuarine sediments, setting up and calibrating a benthic module proves a difficult task. Furthermore, to a certain degree model parameters (such as organic matter degradation and denitrification rate constant) implicitly account for benthic dynamics. We nonetheless acknowledge that, by ignoring benthic processes and burial in particular, our estimates for the estuarine carbon filtering may be underestimated, particularly in the shallow systems of the SAR.***"

In addition, although, the discussion is not centred around the role of benthic processes, a paragraph of the new section 3.4 is also dedicated the difficulty of quantifying the uncertainties of model simulations and an attempt is made using the sensitivity analysis performed by Volta et al. (2014, 2016b).

"***Biogeochemical model parameters for regional and global applications are notoriously difficult to constrain (Volta et al., 2016b). Model parameters implicitly account for processes that are not explicitly resolved and their transferability between systems is thus limited. In addition, published parameter values are generally biased towards temperate regions in industrialized countries (Volta et al., 2016b). A first order estimation of the parameter uncertainty associated to the estuarine carbon removal efficiency (CFilt) can be extrapolated from the extensive parameter sensitivity analyses carried out by Volta et al. (2014, 2016b). These comprehensive sensitivity studies on end-member systems have shown that the relative variation in Cfilt when a number of key biogeochemical parameters are varied by two orders of magnitude varies by is ±15 % in prismatic (short residence time on order of days) to ±25 % in funnel-shaped (long residence time) systems. Thus, assuming that uncertainty increases linearly between those bounds as a function of residence time, an uncertainty estimate can be obtained for each of our modelled estuary. With this simple method, the simulated regional Cfilt of 1.9 Tg C yr^{-1} would be associated with an uncertainty range comprised between 1.5 and 2.2 Tg C yr^{-1}. Our regional estuarine CO_2 evasion estimate is thus reported with moderate confidence. Furthermore, in the future, this uncertainty range could be further constrained using statistical methods such as Monte Carlo simulations (e.g. Lauerwald et al., 2015)."***

L408 Fluxes

Done

L430 Boundary conditions and forcings differ from European settings. Show validations for American estuaries.

We added validations for the American estuaries. Section 2.6 (Model-data comparison) now includes a comparison between model-predicted annual CO_2 outgassing fluxes and 13 published flux estimates, derived from direct measurements in local estuaries to section 2.6 (Model-data comparison). In addition, we provide a validation of our hydrodynamic model using several seasonal longitudinal salinity profiles in the Delaware Bay as well a validation of our biogeochemical model on the basis of pH and pCO_2 profiles from two estuaries (the Delaware Bay and the Altamaha estuary). These additional simulations reveal that C-GEM is able to reproduce observed pCO_2 (Delaware Bay) and both pH and pCO_2 longitudinal profiles along the estuarine gradient (Altamaha).

"***Although C-GEM has been specifically designed and tested for the type of regional application presented here, its transferability from North Sea to US East Coast estuaries was further evaluated by assessing its performance in two East Coast estuaries. First, the hydrodynamic and transport model was tested for the Delaware Bay (MAR). The model was forced with the monthly, minimal and maximal observed discharge at Trenton over the period between 1912 and 1985 (UNH/GRDC Database). Simulated salinity profiles are compared with salinity observations from January, February, May and June (the months with the highest number of data entries), which were extracted from the UNH/GRDC Database. Fig. 6 shows that the model captures both the salinity intrusion length and the overall shape of the salinity profile well. In addition, the performance of the biogeochemical model and specifically its ability to reproduce pH and pCO_2 profiles was evaluated by a model-data comparison for both the Delaware Bay (MAR) in July 2003 and the Altamaha river estuary (SAR) in October 1995. Similar to Volta et al., 2016a, the test systems were chosen due to their contrasting geometries. The Delaware Bay is a marine dominated system characterized by a pronounced funnel shape, while the Altamaha River has a prismatic estuary characteristic of river dominated systems (Jiang et al., 2008). Monthly upstream boundary conditions for nutrients, as well as observed pH data and calculated pCO_2 are extracted from datasets described in (Sharp, 2010) and (Sharp et al., 2009) for the Delaware and in (Cai and Wang, 1998; Jiang et al., 2008) and (Cai et al., 1998) for the Altamaha river estuary. The additional forcings and boundary conditions are set similarly to the simulation for 2000 (see table 2, 3, 4, 5, 6 in SI). Fig. 7 shows that measured and simulated pH values are in good agreement with observed pH and observation-derived calculations of pCO_2. In the Delaware Bay, a pH minimum is located around km 140 and is mainly caused by intense nitrification sustained by large inputs of NH_4 from the Philadelphia urban area, coupled to an intense heterotrophic activity. Both processes lead to a well-developed pCO_2 increase in this area (Fig. 7b). Although no pCO_2 data were available for validation for the period from which boundary conditions were extracted, the simulated profile agree with pCO_2 measurement from July 2013 presented by Joesoef et al. (2015) with pCO_2 values close to equilibrium with the atmosphere in the widest section of the Delaware Bay (close to the estuarine mouth) and values above 1200 µatm at salinities below 5. For the Altamaha river estuary, pH steadily increases from typical river to typical coastal ocean values (Fig. 7b). In addition, both observations and model results reveal that outgassing is very intense in the low-salinity region with more than a 5 fold decrease in pCO_2 between salinity 0 and 5 (Fig 7d).***"

In addition, the new section 3.4 (Scope of applicability and model limitations) critically discusses the difficulties of validating regional/global simulations with local data:

"***The generic nature of the applied model approach and, in particular the application of seasonally/annually averaged or model-deduced boundary conditions renders a direct validation of model results on the basis of local and instantaneous observational data (e.g. longitudinal profiles), which is likely not representative of these long-term average conditions, difficult. Therefore, model performance is evaluated on the basis of spatially aggregated estimates (e.g. regional FCO_2 estimates based on local measurements) rather than system-to-system comparisons with longitudinal profile from specific days. However, note that the performance of C-GEM has been intensively tested by specific model-data comparisons for a number of different systems (e.g. Volta et al., 2014, 2016a) and we are thus confident of its predictive capabilities.***"

L443 a regional minimum

Done

L440-456 Give these numbers in a table and discuss the most relevant ones.

These results were compiled for all estuaries and seasons in supplementary table SI1 and discussed within the text of section 3.1.

L457-462 The percentages should sum up to 100%

When taking all decimals into account, the percentage values do sum up to 100%. We now provide the exact value in the new manuscript.

"In contrast, the 18 MAR estuaries, with their large relative contribution to the total regional estuarine surface area, account for ***as much as 70.1%*** of the total outgassing."

L466 What do you mean with "aspect ratio"?

Aspect ratio refers to the geometry of the estuary (which subsequently affect its biogeochemical behaviour) and more explicitly refers to the ratio between the estuarine width b0 and convergence length b. A wider, funnel shaped estuary whose dynamics are controlled by a strong marine influence while the dynamics in a narrower prismatic estuary is dominated by the river influence (Savenije, 2001).

In the text, the sentence has been modified and the meaning of the term aspect ratio has been clarified:

"The comparatively larger relative contribution of the NAR to the total NEM as compared to the total FCO_2 can be explained by the importance of the specific aspect ratio for NEM. ***A larger ratio of estuarine width b0 and convergence length b corresponds to a more funnel shaped estuary while a low ratio corresponds to a more prismatic geometry (Savenije, 2001; Volta et al., 2014)***"

L479ff Why do the small estuaries show higher mean values?

In large systems, the total outgassing of CO_2 extends over a much larger surface area. In small estuaries, the surface area acts as a limiting factor for the gas exchange with the atmosphere.

L485 Give more details about the assumptions made to calculate the partitioning for

In addition to the reference to Regnier et al. (2013), more details regarding the method used to calculate the respective contributions to the estuarine CO_2 outgasing (NEM, nitrification and riverine oversaturated CO_2) is now provided.

"***Following the approach used in Regnier et al. (2013),*** the contribution of biogeochemical process to *FCO_2* is assessed by evaluating their individual contribution to DIC and ALK changes ***taking into account the local buffering capacity of an ionic solution when TA and DIC are changing due to internal processes, but ignoring advection and mixing (Zeebe and Wolf-Gladrow 2001). In the present study, we quantify the effect of the NEM on the CO_2 balance, which is almost exclusively controlled by aerobic degradation rates because the contributions of denitrification and NPP to the net ecosystem balance are small. Nitrification, a process triggered by the transport and/or production of NH_4 in oxygenated waters, favors outgassing through its effect on pH, which shifts the acid-base equilibrium of carbonate species and increases the CO_2 concentration. The contribution of supersaturated riverine waters to the overall estuarine CO_2 dynamics is calculated as difference between all the other processes creating or consuming CO_2.***"

Fig. 8a. Were seasonal partitioning combined to overall partitioning?

Indeed, the partitioning presented in figure 8a (and figure 8b) are calculated on the basis of the 4 seasonal fluxes for each estuary. The manuscript was edited to clarify this point:

"Fig. 8a presents the contribution of the annually integrated NEM, nitrification and evasion of supersaturated, DIC enriched riverine waters to the total outgassing for each system, as well as for individual regions of the domain. ***The calculation of these annual values is based on the sum of the seasonal fluxes.***"

L489 Give more details about the different partitioning in the different zones here

An entire paragraph following L489 fully describes the partitioning of the 3 drivers of FCO_2 (NEM, nitrification and riverine CO_2) in the 3 different zones (i.e. NAR, MAR, SAR). This paragraph used to begin on L496 of the previous version of the manuscript. We now explain and discuss the regional breakdown earlier in the text and moved the description of the influence of nitrification and NEM on CO_2 outgassing to L 485. Now the discussion of the contributions of NEM, nitrification and riverine CO_2 to FCO2 in each of the 3 sub regions directly follows the sentence in L.489, pointed out by the reviewer.

"Model results reveal that, regionally, the NEM supports about 50% of the estuarine CO_2 outgassing, while nitrification and riverine DIC inputs sustain about 17% and 33% of the CO_2 emissions, respectively. ~~***Nitrification, a process triggered by the transport and/or production of NH_4 in oxygenated waters, favors outgassing through its effect on pH, which shifts the acid-base equilibrium of carbonate species and increases the CO_2 concentration. In addition, the NEM is almost exclusively controlled by aerobic degradation rates because the contribution of denitrification and NPP to the net ecosystem balance is small.***~~ The relative significance of the three processes described above shows important spatial variability..."

L508 Where is Table S1?

Table S1 was uploaded as a supplementary table and a link to download it was included on the page from which the manuscript could be downloaded (below the PDF symbol). Attached to this reply, we provide an archive containing the updated manuscript as well as all the supplementary information

L577 budgets

Done

L630 The normalization of NEM by a Q10 value appears reasonable. The normalization of FCO2 by a Q10 value must be justified. I'm not convinced of the latter normalization.

The rationale for the normalization of FCO_2 by a Q10 value, using the same approach as the one used for NEM, is the fact that, in many systems, NEM and FCO_2 are intimately linked. For instance, Mayer and Eyre (2012) proposed a linear relationship between NEM and FCO2. Applying the same normalization to both NEM and FCO_2 thus allows testing if a similar relationship can be observed along the entire climatic gradient of the US East Coast.

"In this section, we explore the relationships between such simple physical parameters and indicators of the estuarine carbon processing $\overline{NEM}$, $\overline{FCO_2}$ and *CFilt*. In order to account for the effect of temperature on C dynamics, -$\overline{NEM}$ and $\overline{FCO_2}$ are also normalized to the same temperature (arbitrarily chosen to be 0 degree). These normalized values are obtained by dividing -$\overline{NEM}$ and $\overline{FCO_2}$ by a Q_{10} function *f(T)* (see Volta et al., 2014). ***This procedure allows accounting for the exponential increase in the rate of several temperature dependent processes contributing to the NEM (i.e. photosynthesis, organic carbon degradation…). Applying the same normalization to -$\overline{NEM}$ and $\overline{FCO_2}$ is a way of testing how intimately linked NEM and FCO_2 are in estuarine systems. Indeed linear relationships relating one to the other have been reported (Mayer and Eyre, 2012).***"

L660 ff Here it becomes obvious that f(t) cannot be applied to FCO2.

Indeed, we agree with the reviewer that no clear relationship between Q10-normalized FCO_2 and S/Q can be observed over the entire spectrum of values of S/Q that can be found along the east coast of the US. In fact, our results clearly illustrate that a linear regression between FCO_2 / f(T) and S/Q only provides a good fit using estuaries located in the MAR and SAR regions. The small estuaries from the NAR region, characterized by values of S/Q < 3 d m^{-1} display a significantly different behaviour. We think that it is important to point out that small estuaries show a different biogeochemical response and establishing a range of values of S/Q within which Mayer and Eyre's relationship can be reproduced justifies the use of this normalization of FCO_2 by a Q10 . We modified the text to clarify our approach.

"Thus, the well-documented correlation between $\overline{NEM}$ and $\overline{FCO_2}$ (Maher and Eyre, 2012) does not seem to hold for systems with very short residence times. For systems with S/Q > 3 days m^{-1}, we obtain a regression FCO_2 = -0.64 x *NEM* + 5.96 with a r^2 of 0.46, which compares well with the relation FCO_2 = -0.42 x *NEM* + 12 proposed by Maher and Eyre (2012) who used 24 seasonal estimates from small Australian estuaries. ***However, our results suggest that this relationship cannot be extrapolated to small systems such as those located in the NAR.***"

"As a consequence of the distinct behavior of short residence time systems, the coefficient of determination of the best-fitted power law function relating $\overline{FCO_2}$ and S/Q is only significant if NAR systems are excluded ($y = 31.64\ x^{-0.58}$ with a $r^2 = 0.70$). ***This thus suggest that such relationships (as well as that proposed by Maher and Eyre, 2012) cannot be applied to any system but only those for which S/Q>3 day m^{-1}.***"

L668 whom -> who

Done

L677 In this case the assumption of pCO2 in equilibrium with the atmosphere at the lower boundary contradicts the case "still oversaturated waters .."

In our simulations, the seaward boundary is located 20km away from the estuarine mouth and estuarine waters close to the mouths can thus be still oversaturated.

At the beginning of section 2.4.4, we state that '*For each estuary, the downstream boundary is located 20 km beyond the mouth to minimize the bias introduced by the choice of a fixed concentration boundary condition to characterize the ocean water masses (e.g. Regnier et al., 1998).*'

This assumption is also discussed in a paragraph of the new section 3.4:

"***C-GEM places the lower boundary condition 20 km from the estuarine mouth into the coastal ocean and the influence of this boundary condition on simulated biogeochemical dynamics is thus limited. At the lower boundary condition, direct observations for nutrients and oxygen are extracted from databases such as the World Ocean Atlas (Antonov et al., 2014). However, lower boundary conditions for OC and pCO_2 (zero concentration for OC and assumption of pCO_2 equilibrium at the sea side) are simplified. This approach does not allow addressing the additional complexity introduced by biogeochemical dynamics in the estuarine plume (see Arndt et al., 2011). Yet, these dynamics only play a secondary role in the presented study that focuses on the role of the estuarine transition zone in processing terrestrial-derived carbon."***

L682 No link to Fig. 10d ?

We added a reference to Fig. 10d (now Fig.12d), as well as a brief discussion of the non-normalized results to the text.

"***Figure 12d, which reports non-normalized FCO_2 reveals a monotonous increase of FCO_2 with S/Q. This suggests that, unlike the NEM for which the normalization by a temperature function allowed explaining most of the variability; FCO_2 is mostly controlled by the water residence time within the system.*** Discharge is the main FCO_2 driver in riverine dominated systems, while interactions with marshes are driving the outgassing in marine dominated systems surrounded by marshes."

L739 You really mean "prediction"? Not "projection"?

We agree that term projection is better suited and the text was updated accordingly.

"In regions with better data coverage, such as the one investigated here, our study highlights that the regional-scale quantification, attribution, and ***projection*** of estuarine biogeochemical cycling are now at reach."

L740 As your model is rather based on empirical relations than on first principles, I expect that changed systems due to climate shifts and consequences can change your basic relationship. Please include this aspect in a more careful outlook.

We agree with the remark of the reviewer stating that the domain of applicability of the relationship we found between NEM, temperature and the depth normalized estuarine residence is bound within the range of values observed within our study area. Some of these aspects are tackled in the new section 'Scope of applicability and model limitations'.

Additionally, following the reviewer's recommendation, a sentence was added in the outlook section to account for the limitations of the applicability of the relationships we designed. We would like however, to draw the attention of the reviewer on the mechanistic nature of our model. Thus, while the relationships presented in section 3.5 are indeed empirical, they stem from results produced by a model that is actually largely based on first principles.

"In the future, such simple relationships, relying on readily available geometric and hydraulic parameters could be used to quantify carbon processing in areas of the world devoid of direct measurements. ***However, it is important to note that such simple relationships are only valid over the range of boundary conditions and forcings explored and may not be applicable to conditions that fall outside of this range.*** In regions with better data coverage, such as the one investigated here, our study highlights that the regional-scale quantification, attribution, and ***projection*** of estuarine biogeochemical cycling are now at reach."

L1021 7(4), 1271-1295

The reference was updated:

"Volta, C., Arndt, S., Savenije, H. H. G., Laruelle, G. G., and Regnier, P.: C-GEM (v 1.0): a new, cost-efficient biogeochemical model for estuaries and its application to a funnel-shaped system, Geosci. Model Dev., ***7, 1271-1295, doi:10.5194/gmd-7-1271-2014***, 2014."

L1045 give units and if possible your own values.

Following the reviewer's advice, table 3 (now table 2) has been updated to include the unit of the values in the caption and the values calculated by our simulations for the selected estuaries.

"**Table 2**: Published local annually averaged estimates of $\overline{FCO_2}$ ***in mol C m^{-2} yr^{-1}*** for estuaries along the East coast of the US."

Name	**Lon**	**Lat**	$\overline{FCO_2}$ **Observed.**	**Modeled**	**Reference**
Altamaha Sound	-81.3	31.3	32.4	72.7	Jiang et al. (2008)
Bellamy	-70.9	43.2	3.6	3.9	Hunt et al. (2010)
Cocheco	-70.9	43.2	3.1	3.9	Hunt et al. (2010)
Doboy Sound	-81.3	31.4	13.9	25.7	Jiang et al. (2008)
Great Bay	-70.9	43.1	3.6	3.9	Hunt et al. (2011)
Little Bay	-70.9	43.1	2.4	3.9	Hunt et al. (2011)
Oyster Bay	-70.9	43.1	4	3.9	Hunt et al. (2011)
Parker River estuary	-70.8	42.8	1.1	3.9	Raymond and Hopkinson (2003)
Sapelo Sound	-81.3	31.6	13.5	20.6	Jiang et al. (2008)
Satilla River	-81.5	31	42.5	25.7	Cai and Wang (1998)
York River	-76.4	37.2	6.2	8.1	Raymond et al. (2000)
Hudson River	-74	40.6	13.5	15.5	Raymond et al. (1997)
Florida Bay	-80.68	24.96	1.4	n.a.	Dufore (2012)

In addition, the text of the section 2.6 (Model-data comparison) has also been updated to compare observed and simulated FCO_2 in these 13 systems.

"***While such local validations allow assessing the performance of the model for a specific set of conditions, the purpose of this study is to capture the average biogeochemical behaviour of the estuaries of the eastern coast of the US. Therefore, in addition to the system-specific validation, published annually averaged FCO_2 estimates for 13 tidal systems located within the study area collected over the 1994-2006 period are compared to simulated FCO_2 for conditions representative of the year 2000. Overall, simulated FCO_2 are comparable to values reported in the literature (Tab. 2). Although discrepancies, which sometimes can significant, are observed at the level of individual systems, the model captures remarkably well the overall trend in CO_2 evasion rate across estuaries. The model simulates low CO_2 efflux (< 5 mol C m^{-2} yr^{-1}) for the 7 systems were such conditions have been observed, while the 6 systems for which the CO_2 evasion exceeds 10 mol C m^{-2} yr^{-1} are the same in the observations and in the model runs. The discrepancy at the individual system level likely result from a combination of factors, including the choice of model processes and there parametrization, the uncertainties in constraining boundary conditions and the limited representability of instantaneous and local observed.***"

L1052 definition of winter (DJF)?

We define winter in section 2.4 as January, February and March. The definitions of the seasons are now reiterated in the table caption of table 5 to avoid any confusion:

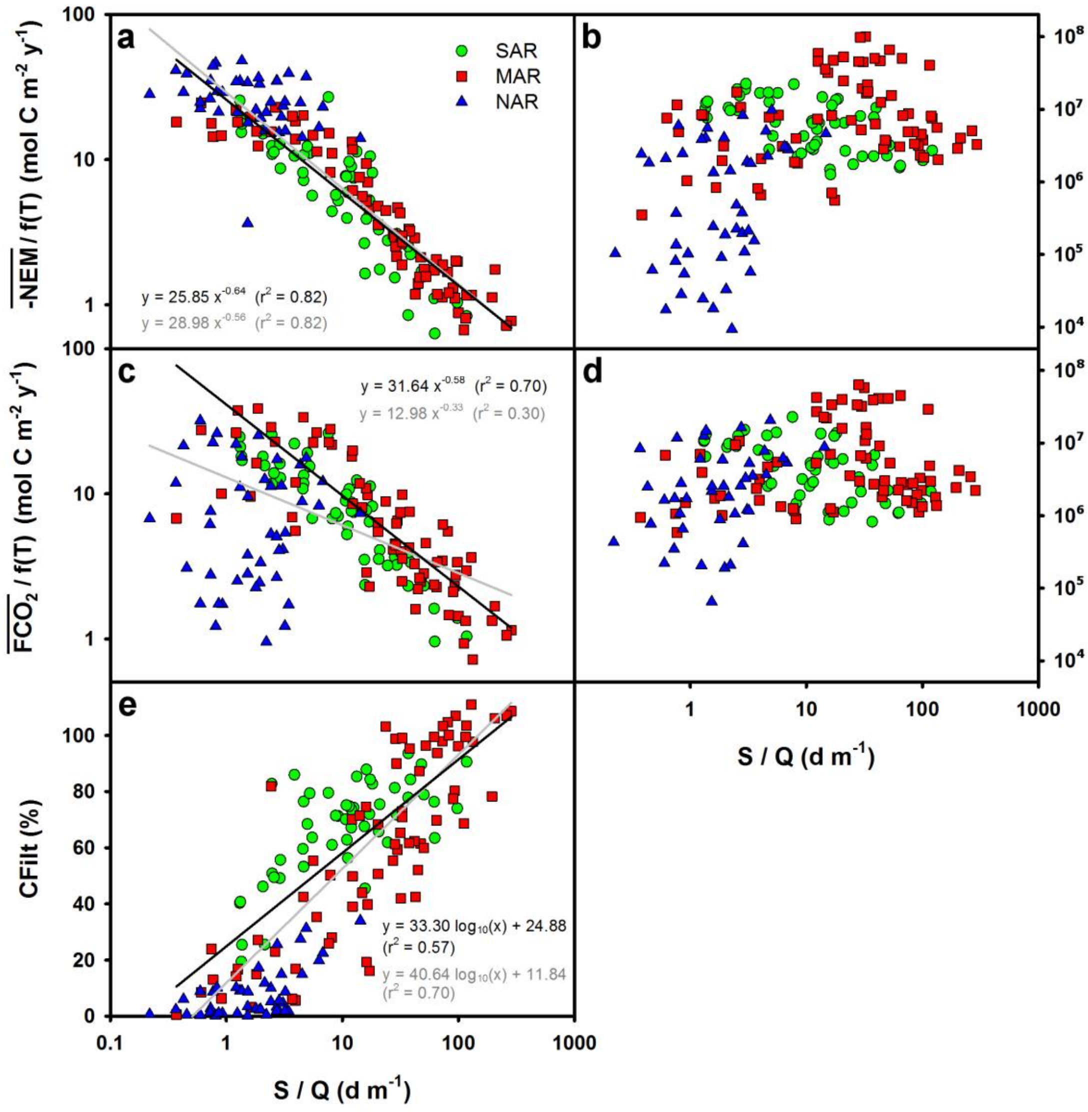
"Table 5: Seasonal contribution to FCO_2 and NEM in each the sub-region. The seasons displaying the highest percentages are indicated in bold. ***Winter is defined as January, February and March, Spring as April, May and June and so on…***"

L1103 The caption must be understandable alone. & L1105 Separate: “black lines .. using all points” “ grey lines are best fit only for ..”

The caption was rewritten taking into account the suggestion of the reviewer:

“Figure 10: ***System scale integrated biogeochemical indicators expressed*** as a function of the depth normalized residence time expressed as the ratio of the estuarine surface S and the river discharge Q ***for all seasons. Panels b, d and e represent NEM, -FCO$_2$ and CFilt, respectively.*** ***Panels a and c represent NEM, -FCO$_2$ normalized by a temperature Q$_{10}$ function.*** Black lines are the best fitted ***linear*** regressions obtained using all the point. ***Grey lines are best fit using only*** the estuaries from the MAR and SAR regions.”

In addition, the y axis of panels b and d were updated.



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Air-water CO_2 evasion from U.S. East Coast estuaries

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Abstract:

This study presents the first regional-scale assessment of estuarine CO_2 evasion along the East coast of the US (25 – 45 °N). The focus is on 43 tidal estuaries, which together drain a catchment of 697000 km^2 or 76 % of the total area within this latitudinal band. The approach is based on the Carbon – Generic Estuarine Model (C-GEM) that allows simulating hydrodynamics, transport and biogeochemistry for a wide range of estuarine systems using readily available geometric parameters and global databases of seasonal climatic, hydraulic, and riverine biogeochemical information. Our simulations, performed using conditions representative of the year 2000, suggest that, together, US East coast estuaries emit 1.9 TgC yr^{-1} in the form of CO_2, which correspond to about 40 % of the carbon inputs from rivers, marshes and mangroves. Carbon removal within estuaries results from a combination of physical (outgassing of supersaturated riverine waters) and biogeochemical processes (net heterotrophy and nitrification). The CO_2 evasion and its underlying drivers show important variations across individual systems, but reveal a clear latitudinal pattern characterized by a decrease in the relative importance of physical over biogeochemical processes along a North-South gradient. Finally, the results reveal that the ratio of estuarine surface area to the river discharge, S/Q (which has a scale of per meter discharged water per year), could be used as a predictor of the estuarine carbon processing in future regional and global scale assessments.

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1 Introduction

Carbon fluxes along the land-ocean aquatic continuum are currently receiving increasing attention because of their recently recognized role in the global carbon cycle and anthropogenic CO_2 budget (Bauer et al., 2013; Regnier et al., 2013a; LeQuéré et al., 2014, 2015). Estuaries are important reactive conduits along this continuum, which links the terrestrial and marine global carbon cycles (Cai, 2011). Large amounts of terrestrial carbon transit through these systems, where they mix with carbon from autochthonous, as well as marine sources. During estuarine transit, heterotrophic processes degrade a fraction of the allochthonous and autochthonous organic carbon inputs, supporting a potentially significant, yet poorly quantified CO_2 evasion flux to the atmosphere. Recent estimates suggest that 0.15-0.25 PgC yr^{-1} is emitted from estuarine systems worldwide (Borges and Abril, 2012; Cai, 2011; Laruelle et al., 2010; Regnier et al., 2013a; Laruelle et al., 2013, Bauer et al., 2013). Thus, in absolute terms the global estuarine CO_2 evasion corresponds to about 15% of the open ocean CO_2 uptake despite the much smaller total surface area.

Currently, estimates of regional and global estuarine CO_2 emissions are mainly derived on the basis of data-driven approaches that rely on the extrapolation of a small number of local measurements (Cai, 2011; Chen et al., 2013; Laruelle et al., 2013). These approaches fail to capture the spatial and temporal heterogeneity of the estuarine environment (Bauer et al., 2013) and are biased towards anthropogenically influenced estuarine systems located in industrialized countries (Regnier et al., 2013a). Even in the best surveyed regions of the world (e.g. Australia, Western Europe, North America or China) observations are merely available for a small number of estuarine systems. In addition, if available, data sets are generally of low spatial and temporal resolution. As a consequence, data-driven approaches can only provide first-order estimates of regional and global estuarine CO_2 emissions.

Integrated model-data approaches can help here, as models provide the means to extrapolate over temporal and spatial scales and allow disentangling the complex and very dynamic network of

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physical and biogeochemical processes that controls estuarine CO_2 emissions. Over the past decades, increasingly complex process-based models have been applied, in combination with local data, to elucidate the coupled carbon-nutrient cycles on the scale of individual estuaries (e.g., O'Kane, 1980; Soetaert and Herman, 1995; Vanderborght et al., 2002; Lin et al., 2007; Arndt et al., 2009; Cerco et al., 2010; Baklouti et al., 2011). However, the application of such model approaches remains limited to the local scale due to their high data requirements for calibration and validation (e.g. bathymetric and geometric information and boundary conditions), as well as the high computational demand associated with resolving the complex interplay of physical and biogeochemical processes on the relevant temporal and spatial scales (Regnier et al., 2013b). Complex process-based models are thus not suitable for the application on a regional or global scale and, as a consequence, the estuarine carbon filter is, despite its increasingly recognized role in regional and global carbon cycling (e.g. Bauer et al., 2013), typically not taken into account in model-derived regional or global carbon budgets (Bauer et al., 2013). The lack of regional and global model approaches that could be used as stand-alone applications or that could be coupled to regional terrestrial river network models (e.g. GLOBALNEWS: Seitzinger et al., 2005; Mayorga et al., 2010; SPARROW: Schwarz et al., 2006) and continental shelf models (e.g. Hofmann et al., 2011) is thus critical.

The Carbon-Generic Estuary Model (C-GEM (v1.0); Volta et al., 2014) has been developed with the aim of providing such a regional/global modeling tool that can help improve existing, observationally derived first order estimates of estuarine CO_2 emissions. C-GEM (v1.0) has been specifically designed to reduce data requirements and computational demand and, thus, tackles the main impediments for the application of estuarine models on a regional or global scale. The approach takes advantage of the mutual dependency between estuarine geometry and hydrodynamics in alluvial estuaries and uses an idealized representation of the estuarine geometry to support the hydrodynamic calculations. It thus allows running steady state or fully transient annual to multi-decadal simulations for a large number of estuarine systems, using geometric information readily available through maps

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or remote sensing images. Although the development of such a regional/global tool inevitably requires simplification, careful model evaluations have shown that, despite the geometric simplification, C-GEM provides an accurate description of the hydrodynamics, transport and biogeochemistry in tidal estuaries (Volta et al., 2014). In addition, the model approach was successfully used to quantify the contribution of different biogeochemical processes for CO_2 air-water fluxes in an idealized, funnel-shaped estuary forced by typical summer conditions characterizing a temperate Western European climate (Regnier et al., 2013b). Volta et al. (2016b) further investigated the effect of estuarine geometry on the CO_2 outgassing using three idealized systems and subsequently established the first regional carbon budget for estuaries surrounding the North Sea by explicitly simulating the six largest systems of the area (Volta et al., 2016a), including the Scheldt and the Elbe for which detailed validation was performed.

Here, we extend the domain of application of C-GEM (v1.0) to quantify CO_2 exchange fluxes, as well as the overall organic and inorganic carbon budgets for the full suite of estuarine systems located along the entire East coast of the United States, one of the most intensively monitored regions in the world. A unique set of regional data, including partial pressure of CO_2 in riverine and continental shelf waters (pCO_2; Signorini et al., 2013; Laruelle et al., 2015), riverine biogeochemical characteristics (Lauerwald et al., 2013), estuarine eutrophication status (Bricker et al., 2007) and estuarine morphology (NOAA, 1985) are available. These comprehensive data sets are complemented by local observations of carbon cycling and CO_2 fluxes in selected, individual estuarine systems (see Laruelle et al., 2013 for a review), making the East coast of the United States an ideal region for a first, fully explicit regional evaluation of CO_2 evasion resolving every major tidal estuary along the selected coastal segment. The scale addressed in the present study is unprecedented so far (> 3000 km of coastline) and covers a wide range of estuarine morphological features, climatic conditions, land-use and land cover types, as well as urbanization levels. The presented study will not only allow a further evaluation of C-GEM (v1.0), but will also provide the first regional-scale assessment of estuarine CO_2 evasion along the East coast of the US (25 – 45 °N)

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and will help explore general relationships between carbon cycling and CO_2 evasion, and readily available estuarine geometrical parameters.

After a description of the model itself and of the dataset used to set up the simulations, a local validation is presented which includes salinity, pCO_2 and pH longitudinal profiles for two well monitored systems (the Delaware Bay and the Altamaha River Estuary). The yearly averaged rates of CO_2 exchange at the air-water interface simulated by the model for 13 individual estuaries are also compared with observed values reported in the literature. Next, regional scale simulations for 43 tidal estuaries of the eastern US coast provide seasonal and yearly integrated estimates of the Net Ecosystem Metabolism (NEM), CO_2 evasion and carbon filtering capacity, CFilt. Model results are then used to elucidate the estuarine biogeochemical behavior along the latitudinal transect encompassed by the present study (30-45° N). Finally, our results are used to derive general relationships between carbon cycling and CO_2 evasion, and readily available estuarine geometrical parameters.

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2. Regional description and model approach

2.1 Observation-based carbon budget for the East coast of the United States

The study area covers the Atlantic coast of the United States (Fig.1), from the southern tip of Florida (25°N) to Cobscook Bay (45°N) at the US-Canada boundary. This area encompasses distinct climatic zones and land cover types and exhibits a variety of morphologic features (Fig. 1). The region can be subdivided into several sub-regions following a latitudinal gradient (Signorini et al., 2013). In this study, we define three sub-regions following the boundaries suggested by the COSCAT segmentation (Meybeck et al., 2006; Laruelle et al., 2013) and the further subdivision described in Laruelle et al. (2015). From North to South, the regions are called North Atlantic, Mid Atlantic and South Atlantic Regions (Fig. 1). Total carbon inputs from watersheds to US East coast estuaries (Tab. 1) have been

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estimated to range from 4.0 to 10.7 Tg C yr^{-1} (Mayorga et al., 2010; Shih et al., 2010; Stets and Strieg, 2012; Tian et al., 2010; Tian et al., 2012), consisting of dissolved organic carbon (DOC; ~50%), dissolved inorganic carbon (DIC; ~40%) and particulate organic carbon (POC; ~10%). In addition, a statistical approach has been applied to estuaries of the region to quantify organic carbon budgets and Net Ecosystem Productivity (NEP) using empirical models (Herrmann et al., 2015).

Recent studies estimated that, along the East coast of the United States, rivers emit 11.4 TgC yr^{-1} of CO_2 to the atmosphere (Raymond et al., 2013), while continental shelf waters absorb between 3.4 and 5.4 TgC yr^{-1} of CO_2 from the atmosphere (Signorini et al., 2013). A total of thirteen local, annual mean estuarine CO_2 flux estimates across the air-water interface based on measurements are also reported in the literature and are grouped along a latitudinal gradient (Tab. 2). Four of these estimates are located in the South Atlantic region (SAR): Sapelo Sound, Doboy Sound, Altamaha Sound (Jiang et al., 2008), and the Satilla River estuary (Cai and Wang, 1998). Three studies investigate CO_2 fluxes in the mid-Atlantic Region (MAR): the York River Estuary (Raymond et al., 2000) and the Hudson River (Raymond et al., 1997). There is also a comprehensive CO_2 flux study for the Delaware Estuary published after the completion of this work (Joeseof et al., 2015). Six systems are located in the North Atlantic region (NAR): The Great Bay, the Little Bay, the Oyster estuary, the Bellamy estuary, the Cocheco estuary (Hunt et al., 2010; 2011), and the Parker River estuary (Raymond and Hopkinson, 2003). The mean annual flux per unit area from these local studies is 11.7±13.1 mol C m^{-2} yr^{-1} and its extrapolation to the total estuarine surface leads to a regional CO_2 evasion estimate of 3.8 Tg C y^{-1}. This estimate is in line with that of Laruelle et al. (2013) for the same region which proposes an average CO_2 emission rate of 10.8 mol C m^{-2} yr^{-1}. Thus, CO_2 outgassing could remove 35% to 95% of the riverine carbon loads during estuarine transit. About 75 % of the air-water exchange occurs in tidal estuaries (2.8 Tg C y^{-1}) while lagoons and small deltas contribute to the remaining 25 %. Although these simple extrapolations from limited observational data are associated with large uncertainties, they highlight the potentially significant contribution of estuaries to the CO_2 outgassing in the region. However, process-based quantifications of regional organic and

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inorganic C budgets including air-water CO_2 fluxes for the estuarine systems along the East coast are not available.

2.2 Selection of estuaries

The National Estuarine Eutrophication Assessment (NEAA) survey (Bricker et al., 2007), which uses geospatial data from the National Oceanic and Atmospheric Administration (NOAA) Coastal Assessment Framework (CAF) (NOAA, 1985), was used to identify and characterize 58 estuarine systems discharging along the Atlantic coast of the United States. From this set, 43 'tidal' estuaries, defined as a river stretch of water that is tidally influenced (Dürr et al., 2011), were retained (Fig. 1) to be simulated by the C-GEM model, which is designed to represent such systems. Using outputs from terrestrial models (Hartmann et al., 2009; Mayorga et al., 2010), the cumulated riverine carbon loads for all the non-tidal estuaries that are excluded from the present study amount to 0.9 Tg C yr^{-1}, which represents less than 15% of the total riverine carbon loads of the region. These 15 systems are located in the SAR (10) and in the MAR (5).

The northeastern part of the domain (NAR, Fig. 1; Tab. 1) includes 20 estuaries along the Gulf of Maine and the Scotian shelf, covering a cumulative surface area of ~5300 km². It includes drowned valleys, rocky shores and a few tidal marshes. The climate is relatively cold (annual mean= 8°C) and the human influence is relatively limited because of low population density and low freshwater inputs. The mean estuarine water depth is 12.9 m and the mean tidal range is 2.8 m.

The central zone (MAR) includes 17 tidal estuaries accounting for a total surface area of 14500 km². The Chesapeake Bay and the Delaware estuaries alone contribute more than 60% to the surface area of the region. In this region, estuaries are drowned valleys with comparatively high river discharge and intense exchange with the ocean. Several coastal lagoons, characterized by a limited exchange with the ocean are located here, but are not included in our analysis. The Mid-Atlantic Region (MAR) is characterized by a mean annual temperature of 13°C and is strongly impacted by human activities,

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due to the presence of several large cities (e.g. New York, Washington, Philadelphia, Baltimore) and intense agriculture. The mean water depth is about 4.7 m and the tidal range is 0.8 m.

The southern Atlantic region (SAR) includes 10 tidal estuaries covering a total surface area of 12182 km². These systems are generally dendritic and surrounded by extensive salt marshes. The climate is subtropical with an average annual temperature of 19°C. Land use includes agriculture and industry, but the population density is generally low. Estuarine systems in the SAR are characterized by a shallow mean water depth of 2.9 m and a tidal range of 1.2 m.

2.3 Model set-up

The generic 1D Reactive-Transport Model (RTM) C-GEM (Volta et al., 2014) is used to quantify the estuarine carbon cycling in the 43 systems considered in this study. The approach is based on idealized geometries (Savenije, 2005; Volta et al., 2014) and is designed for regional and global scale applications (Regnier et al., 2013b; Volta et al., 2014, 2016a). The model approach builds on the premise that hydrodynamics exerts a first-order control on estuarine biogeochemistry (Arndt et al., 2007; Friedrichs and Hofmann, 2001) and CO_2 fluxes (Regnier et al., 2013a). The method takes advantage of the mutual dependence between geometry and hydrodynamics in tidal estuaries (Savenije, 1992) and the fact that, as a consequence, transport and mixing can be easily quantified from readily available geometric data (Regnier et al., 2013a; Savenije, 2005; Volta et al., 2016b).

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2.3.1 Description of idealized geometries for tidally-averaged conditions

Although tidal estuaries display a wide variety of shapes, they nevertheless share common geometric characteristics that are compatible with an idealized representation (Fig. 2, Savenije, 1986; Savenije, 2005). For tidally-averaged conditions, their width B (or cross-sectional area A) can be described by an exponential decrease as a function of distance, *x*, from the mouth (Savenije, 1986; Savenije, 2005):

$$B = B0 * \exp\left(-\frac{x}{b}\right) \quad (1)$$

where B (m) is the tidally averaged width, B0 (m) the width at the mouth, x (m) the distance from the mouth (x=0) and b (m) the width convergence length (Fig. 2). The width convergence length, b, is defined as the distance between the mouth and the point at which the width is reduced to B0 e^{-1}. It is directly related to the dominant hydrodynamic forcing. A high river discharge typically results in a prismatic channel with long convergence length (river dominated estuary), while a large tidal range results in a funnel-shaped estuary with short convergence length (marine dominated estuary). At the upstream boundary, the estuarine width is given by:

$$B_L = B0 * \exp\left(-\frac{L}{b}\right) \quad (2)$$

Where L denotes the total estuarine length (m) along the estuarine longitudinal axis.

The total estuarine surface S (m^2) can be estimated by integrating equation (1) over the estuarine length:

$$S = \int_0^L B\,dx = b * B0 * \left(1 - \exp\left(-\frac{L}{b}\right)\right) \quad (3)$$

The width convergence length is then calculated from B0, B_L, L and the real estuarine surface area (SR) by inserting equation (2) in equation (3):

$$b = \frac{SR}{B0 - BL} \quad (4)$$

SR is calculated for each system using the SRTM water body data (Fig. 3a), a geographical dataset encoding high-resolution worldwide coastal outlines in a vector format (NASA/NGA, 2003). While such a database exists for a well monitored region such as the East coast of the US, resorting to using the idealized estuarine surface area (S) is necessary in many other regions. The longitudinal

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mean, tidally averaged, depth h (m), is obtained from the National Estuarine Eutrophication Assessment database (Bricker et al., 2007).

Using this idealized representation, the estuarine geometry can be defined by a limited number of parameters: the width at the mouth (B_0), the estuarine length (L), the estuarine width at the upstream limit (B_L) and the mean depth h. These parameters can be easily determined from local maps or Google Earth using Geographic Information Systems (GIS) or obtained from databases (NASA/NGA, 2003).

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2.3.2 Hydrodynamics, transport and biogeochemistry

Estuarine hydrodynamics are described by the one-dimensional barotropic, cross-sectionally integrated mass and momentum conservation equations for a channel with arbitrary geometry (Nihoul and Ronday, 1976; Regnier et al., 1998; Regnier and Steefel, 1999):

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$$r_s \frac{\partial A}{\partial t} + \frac{\partial Q}{\partial x} = 0 \qquad (5)$$

$$\frac{\partial U}{\partial t} + U \frac{\partial U}{\partial x} = -g \frac{\partial \zeta}{\partial x} - g \frac{U|U|}{C_z^2 H} \qquad (6)$$

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where:

t	time	[s]
x	distance along the longitudinal axis	[m]
A	cross-section area $A = H \cdot B$	[m^2]
Q	cross-sectional discharge $Q = A \cdot U$	[$m^3\ s^{-1}$]
U	flow velocity Q / A	[$m\ s^{-1}$]
r_s	storage ratio $r_s = B_s / B$	[-]

B_s	storage width	[m]
g	gravitational acceleration	[m s^{-2}]
ξ	elevation	[m]
H	total water depth $H = h + \xi(x,t)$	[m]
C_z	Chézy coefficient	[m$^{1/2}$ s^{-1}]

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The coupled partial differential equations (Eqs. (5) and (6)) are solved by specifying the elevation ξ_0(t) at the estuarine mouth and the river discharge Q_r(t) at the upstream limit of the model domain.

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The one-dimensional, tidally-resolved, advection-dispersion equation for a constituent of concentration *C(x,t)* in an estuary can be written as (e.g. Pritchard, 1958):

$$\frac{\partial C}{\partial t} + \frac{Q}{A}\frac{\partial C}{\partial x} = \frac{1}{A}\frac{\partial}{\partial x}\left(AD\frac{\partial C}{\partial x}\right) + P \qquad (7)$$

where *Q*(x,t) and *A*(x,t) denote the cross-sectional discharge and area, respectively and are provided by the hydrodynamic model (eq. 5 and 6). *P*(x,t) is the sum of all production and consumption process rates affection the concentration of the constituent. The effective dispersion coefficient *D* (m^2 s^{-1}) implicitly accounts for dispersion mechanisms associated to sub-grid scale processes (Fischer, 1976; Regnier et al., 1998). In general, *D* is maximal near the sea, decreases upstream and becomes virtually zero near the tail of the salt intrusion curve (Preddy, 1954; Kent, 1958; Ippen and Harleman, 1961; Stigter and Siemons, 1967). The effective dispersion at the estuarine mouth can be quantified by the following relation (Savenije, 1986):

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$$D_0 = 26 \cdot (h_0)^{1.5} \cdot (N \cdot g)^{0.5} \qquad (8)$$

where h_0 (m) is the tidally-averaged water depth at the estuarine mouth and *N* is the dimensionless Canter Cremers' estuary number defined as the ratio of the freshwater entering the estuary during a tidal cycle to the volume of salt water entering the estuary over a tidal cycle (Simmons, 1955).

$$N = \frac{Q_b \cdot T}{P} \qquad (9)$$

In this equation, Q_b is the bankfull discharge ($m^3\ s^{-1}$), T is the tidal period (s) and P is the tidal prism (m^3). For each estuary, N can thus be calculated directly from the hydrodynamic model. The variation in *D* along the estuarine gradient can be described by Van der Burgh's equation (Savenije, 1986):

$$\frac{\partial D}{\partial x} = -K\frac{Q_r}{A} \qquad (10)$$

where *K* is the dimensionless Van der Burgh's coefficient and the minus sign indicates that *D* increases in downstream direction (Savenije, 2012). The Van der Burgh's coefficient is a shape factor that has values between 0 and 1 (Savenije, 2012), and is a function of estuarine geometry for tidally average conditions. Therefore, each estuarine system has its own characteristic *K* value, which correlates with geometric and hydraulic scales (Savenije, 2005). Based on a regression analysis covering a set of 15 estuaries, it has been proposed to constrain *K* from the estuarine geometry (Savenije, 1992):

$$K = 4.32 \cdot \frac{h_0^{0.36}}{B_0^{0.21} \cdot b^{0.14}} \quad \text{with} \quad 0 < K < 1 \qquad (11)$$

Reaction processes *P* considered in C-GEM comprise aerobic degradation, denitrification, nitrification, primary production, phytoplankton mortality and air-water gas exchange for O_2 and CO_2 (Fig. 4 and Tab. 3). These processes and their mathematical formulation are described in detail in Volta et al. (2014) and Volta et al. (2016a).

The non-linear partial differential equations for the hydrodynamics are solved by a finite difference scheme following the approach of (Regnier et al., 1997; Regnier and Steefel, 1999) and (Vanderborght et al., 2002). The timestep Δt is 150s and the grid size Δx is constant along the

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longitudinal axis of the estuary. The grid size default value is 2000 m, but can be smaller for short length estuaries to guarantee a minimum of 20 grid points within the computational domain. Transport and reaction terms are solved in sequence within a single timestep using an operator splitting approach (Regnier et al., 1997). The advection term in the transport equation is integrated using a third-order accurate total variation diminishing (TVD) algorithm with flux limiters (Regnier et al., 1998), ensuring monotonicity (Leonard, 1984), while a semi-implicit Crank-Nicholson algorithm is used for the dispersion term (Press et al., 1992). These schemes have been extensively tested using the CONTRASTE estuarine model (e.g. Regnier et al., 1998; Regnier and Steefel, 1999; Vanderborght et al., 2002) and guarantee mass conservation to within <1%. The reaction network (including erosion-deposition terms when the constituent is a solid species), is numerically integrated using the Euler method (Press et al., 1992). The primary production dynamics, which requires vertical resolution of the photic depth, is calculated according to the method described in Vanderborght et al. (2007). This method assumes an exponential decrease of the light in the water column (Platt et al., 1980), which is solved using a Gamma function.

2.4 Boundary and forcing conditions

Boundary and forcing conditions are extracted from global databases and global model outputs that are available at 0.5° resolution. Therefore, C-GEM simulations are performed at the same resolution according to the following procedure. First, 43 coastal cells corresponding to tidal estuaries are identified in the studied area (Fig. 1). If the mouth of an estuary is spread over several 0.5° grid cells, those cells are regrouped in order to represent a single estuary (e.g. Delaware estuary), and subsequently, a single idealized geometry is defined as described above. The model outputs (Hartmann et al. , 2009; Mayorga et al., 2010) and databases (Antonov et al., 2010; Garcia et al., 2010a; Garcia et al., 2010b) used to constrain our boundary conditions are representative of the year 2000.

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For each resulting cell, boundary and forcing conditions are calculated for the following periods: January-March; April-June; July-September and October-December. This allows for an explicit representation of the seasonal variability in the simulations.

2.4.1 External forcings

Transient physical forcings are calculated for each season and grid cell using monthly mean values of water temperature (World Ocean Atlas, 2009) and seasonal averaged values for wind speed (Cross-Calibrated-Multi-Platform (CCMP) Ocean Surface Wind Vector Analyses project (Atlas et al., 2011)). Mean daily solar radiation and photoperiods (corrected for cloud coverage using the ISCCP Cloud Data Products, Rossow and Schiffer, 1999) are calculated depending on latitude and day of the year using a simple model (Brock, 1981).

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2.4.2 Riverine discharge, concentrations and fluxes

River discharges are extracted from the UNH/GRDC runoff dataset (Fekete et al., 2002). These discharges represent long-term averages (1960-1990) of monthly and annual runoff at 0.5 degree resolution. The dataset is a composite of long-term gauging data, which provides average runoff for the largest river basins, and a climate driven water balance model (Fekete et al., 2002). Total runoff values are then aggregated for each watershed at the coarser 0.5 degree resolution (Fig. 3b). Next, seasonal mean values (in m^3 s^{-1}) are derived in order to account for the intra-annual variability in water fluxes. Based on annual carbon and nutrients inputs from the watersheds (Mg y^{-1}), mean annual concentrations (mmol m^{-3}) are estimated for each watershed using the UNH/GRDC annual runoff (km^3 y^{-1}). Mean seasonal concentrations are then calculated from the seasonally resolved river water fluxes of a given sub-region.

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Annual inputs of dissolved organic carbon (DOC), particulate organic carbon (POC) and inorganic nutrients are derived from the globalNEWS2 model (Mayorga et al., 2010). Global NEWS is a spatially explicit, multi-element (N, P, Si, C) and multi-form global model of nutrient exports by rivers. In a nutshell, DOC exports are a function of runoff, wetland area, and consumptive water use (Harrison et al., 2005). No distinction is made between agricultural and natural landscapes, since they appear to have similar DOC export coefficients (Harrison et al., 2005). Sewage inputs of OC are ignored in GlobalNEWS, because their inclusion did not improve model fit to data (Harrison et al., 2005). POC exports from watersheds are estimated using an empirical relationship with Suspended Particulate Matter (SPM; Ludwig et al., 1996). Inorganic nitrogen (DIN) and phosphorus (DIP) fluxes calculated by GlobalNEWS depend on agriculture and tropical forest coverage, fertilizer application, animal grazing, sewage input, atmospheric N deposition and biological N fixation (Mayorga et al., 2010). The inputs of dissolved silica (DSi) are controlled by soil bulk density, precipitation, slope, and presence of volcanic lithology (Beusen et al., 2009).

The DIN speciation is not provided by the GlobalNEWS2 model. The NH_4 and NO_3 concentrations are therefore determined independently on the basis of an empirical relationship between ammonium fraction (NH4/DIN ratio) and DIN loads (Meybeck, 1982). Dissolved Oxygen (DO) concentrations are extracted from the water quality criteria recommendations published by the United States Environmental Protection Agency (EPA, 2009). The same source is used for phytoplankton concentrations, using a chlorophyll-a to phytoplankton carbon ratio of 50 gC $(gChla)^{-1}$ (Riemann et al., 1989) to convert the EPA values to carbon units used in the present study.

Inputs of dissolved inorganic carbon (DIC) and total Alkalinity (ALK) are calculated from values reported in the GLORICH database (Hartmann et al., 2009). For each watershed, seasonal mean values of DIC and ALK concentrations are estimated from measurements performed at the sampling locations that are closest to the river-estuary boundary. The spatial distribution of annual inputs of TOC=DOC+POC, DIC, and TC=TOC+DIC from continental watersheds to estuaries are reported in Fig. 5a, 5c and 5d, respectively. The contribution of tidal wetlands to the TOC inputs is also shown (Fig. 5b). Overall, the TC input over the entire model domain is estimated at 4.6 Tg C yr^{-1}, which falls in the lower end of previous reported estimations (Najjar et al. 2012).

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2.4.3 Inputs from tidal wetlands

The DOC input of estuarine wetlands (Fig. 5b) scales to their fraction, W, of the total estuarine and is calculated using the GlobalNEWS parameterization:

$$Y\_DOC = \frac{\left[(E\_C_{wet} * W) + E\_C_{dry} * (1 - W)\right] * R^{a} * Q_{act}}{Q_{nat}} \qquad (12)$$

$$\frac{Y\_DOC_{wet}}{Y\_DOC} = \frac{E\_C_{wet} * W}{E\_C_{wet} * W + E\_C_{dry} * (1 - W)} \qquad (13)$$

where Y_DOC is the DOC yield (kg C km^{-2} y^{-1}) calculated for the entire watershed, Y_DOC_{wet} is the estimated DOC yield from wetland areas (kg C km^{-2} y^{-1}), Q_{act}/Q_{nat} is the ratio between the measured discharge after dam construction and before dam construction, E_C_{wet} and E_C_{dry} (kg C km^{-2} y^{-1}) are the export coefficients of DOC from wetland and non-wetland soils, respectively. W is the percentage of the land area within a watershed that is covered by wetlands, R is the runoff (m y^{-1}) and *a* is a unit-less calibration coefficient defining how non-point source DOC export responds to runoff. The value of a is set to 0.95, consistent with the original GlobalNEWS -DOC model of Harrison et al. (2005). The carbon load Y_DOC_{wet} is then exported as a diffuse source along the relevant portions of estuary. The estuarine segments receiving carbon inputs from tidal wetlands are identified using the National Wetlands Inventory of the U.S. Fish and Wildlife Service (U.S. Fish and Wildlife Service, 2014). The inputs from those systems are then allocated to the appropriate grid cell of the model domain using GIS. The flux calculated is an annual average that is subsequently partitioned between the four seasons as a function of the mean seasonal temperature, assumed to be the main control of the wetland-estuarine exchange. This procedure reflects the observation that

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in spring and early summer, DOC export is small as a result of its accumulation in the salt marshes induced by the high productivity (Dai and Wiegert, 1996), (Jiang et al., 2008). In late summer and fall, the higher water temperature and greater availability of labile DOC contribute to higher bacterial remineralization rates in the intertidal marshes (Cai et al., 1999; Middelburg et al., 1996; Wang and Cai, 2004), which induce an important export. This marsh production-recycle-export pattern is consistent with the observed excess DIC signal in the offshore water (Jiang et al. 2013). DIC export from tidal wetlands is neglected here because it is assumed that OC is not degraded before reaching the estuarine realm. Although this assumption may lead to an overestimation of OC export from marshes and respiration in estuarine water, it will not significantly affect the water pCO_2 and degassing in the estuarine waters because mixing is faster than respiration.

2.4.4 Concentrations at the estuarine mouth

For each estuary, the downstream boundary is located 20 km beyond the mouth to minimize the bias introduced by the choice of a fixed concentration boundary condition to characterize the ocean water masses (e.g. Regnier et al., 1998). This approach also reduces the influence of marine boundary conditions on the simulated estuarine dynamics, especially for all organic carbon species whose concentrations are fixed at zero at the marine boundary. This assumption ignores the intrusion of marine organic carbon into the estuary during the tidal cycle but allows focusing on the fate of terrigenous material and its transit through the estuarine filter. DIC concentrations are extracted from the GLODAP dataset (Key et al., 2004), from which ALK and pH are calculated assuming CO_2 equilibrium between coastal waters and the atmosphere. The equilibrium value is computed using temperature (WOA2009, Locarnini et al., 2010) and salinity (WOA2009, Antonov et al. (2010)) data which vary both spatially and temporally. The equilibrium approach is a reasonable assumption because differences in partial pressure ΔpCO_2 between coastal waters and the atmosphere are generally much smaller (0-250 µatm (Signorini et al., 2013)) than those reported for estuaries (ΔpCO_2 in the range 0-10000 µatm (Borges and Abril, 2012)). Salinity, DO, NO_3, DIP and DSi

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concentrations are derived from the World Ocean Atlas (Antonov et al., 2010; Garcia et al., 2010a; Garcia et al., 2010b). NH_4 concentrations are set to zero in marine waters. For all variables, seasonal means are calculated for each grid cell of the boundary.

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2.5 Biogeochemical indicators

The model outputs (longitudinal profiles of concentration and reaction rates) are integrated in time over the entire volume or surface of each estuary to produce the following indicators of the estuarine biogeochemical functioning (Regnier et al., 2013b): the mean annual Net Ecosystem Metabolism (*NEM*), the air-water CO_2 flux (FCO_2), the carbon and nitrogen filtering capacity (*CFilt* and *NFilt*) and their corresponding element budgets. The *NEM* (molC y^{-1}) (Caffrey, 2004; Odum, 1956) is defined as the difference between net primary production (*NPP*) and total heterotrophic respiration (*HR*) at the system scale:

$$NEM = \int_0^{365} \int_0^{L} [NPP(x,t) - R_{aer}(x,t) - R_{den}(x,t)] * B(x) * H(x,t)\, dx\, dt \qquad (14)$$

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where *NPP* is the Net Primary Production (mol C m^{-3} y^{-1}), R_{aer} the aerobic degradation of organic matter (in mol C m^{-3} y^{-1}) and R_{den} the denitrification (in mol C m^{-3} y^{-1}) (see Volta et al., 2014 for detailed formulations). *NEM* is thus controlled by the production and decomposition of autochthonous organic matter, by the amount and degradability of organic carbon delivered by rivers and tidal wetlands and by the export of terrestrial and in-situ produced organic matter to the adjacent coastal zone. Following the definition of *NEM*, the trophic status of estuaries can be net heterotrophic (NEM<0) when *HR* exceeds *NPP* or net autotrophic (NEM>0), when *NPP* is larger than *HR* because the burial and export of autochthonous organic matter exceeds the decomposition of river-borne material.

The FCO_2 (mol C y^{-1}) is defined as:

$$FCO_2 = \int_0^{365} \int_0^{L} RCO_2(x,t) * B(x)\, dx\, dt \qquad (15)$$

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$$RCO_2(x,t) = -v_p(x,t)\left(\left[CO_{2(aq)}\right](x,t) - K_0(x,t) * P_{CO2}(x,t)\right) \qquad (16)$$

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where RCO_2 (molC m^{-2} y^{-1}) is the rate of exchange in CO_2 at the air-water interface per unit surface area, v_p is the piston velocity (m y^{-1}) and is calculated according to Regnier et al. (2002) to account for the effect of current velocity and wind speed, [*CO2(aq)*] is the concentration of CO_2 in the estuary (mol m^{-3}), K_0 is Henry's constant of CO_2 in sea water (mol m^{-3} atm^{-1}) and P_{CO2} is the atmospheric partial pressure in CO_2 (atm).

The carbon filtering capacity (in %) corresponds to the fraction of the river-borne supply that is lost to the atmosphere and is defined here as the ratio of the net outgassing flux of CO_2 and the total inputs of C, e.g. total carbon expressed as the sum of inorganic and organic carbon species, both in the dissolved and particulate phases.

$$CFilt = \frac{FCO_2}{\int_0^{365} Q * [TC]_{riv}\, dt} * 100 \qquad (17)$$

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where $[TC]_{riv}$ denote the total concentrations of C in the riverine inputs.

Fluxes per unit area for FCO_2 and *NEM*, noted $\overline{FCO_2}$ and $\overline{NEM}$, respectively, are defined in mol C m^{-2} y^{-1} and are calculated by dividing the integrated values calculated above by the (idealized) estuarine surface *S*:

$$\overline{NEM} = \frac{NEM}{S} * 1000 \qquad (18)$$

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$$\overline{FCO_2} = \frac{FCO_2}{S} * 1000 \qquad (19)$$

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Seasonal values for the biogeochemical indicators are calculated using the same formula as above, but calculate the integral over a seasonal rather than annual timescale (i.e. 3 months).

2.6 Model-data comparison

C-GEM has been specifically designed for an application on a global/regional scale requiring the representation of a large number of individual and often data-poor systems. Maximum model transferability and minimum validation requirements were thus central to the model design process and the ability of the underlying approach in reproducing observed dynamics with minimal calibration effort has been extensively tested. The performance C-GEM's one-dimensional hydrodynamic and transport models using idealized geometries have been evaluated for a number of estuarine systems exhibiting a wide variety of shapes (Savenije, 2012). In particular, it has been shown that the estuarine salt intrusion can be successfully reproduced using the proposed modeling approach (Savenije 2005; Volta et al., 2014; 2016b). In addition, C-GEM's biogeochemistry has also been carefully validated for geometrically contrasting estuarine system in temperate climate zones. Simulations for the Scheldt Estuary (Belgium and the Netherlands), a typical funnel-shaped estuary, were validated through model-data and model-model comparison (Volta et al., 2014; Volta et al., 2016a). Furthermore, simulations for the Elbe estuary (Germany), a typical prismatic shape estuary that drains carbonate terrains and, thus, exhibits very high pH was validated against field data (Volta et al., 2016a). In addition, C-GEM carbon budgets have been compared budget derived from observations for 6 European estuaries discharging in the North Sea (Volta et al., 2016a). Although C-GEM has been specifically designed and tested for the type of regional application presented here, its transferability from North Sea to US East Coast estuaries was further evaluated by assessing its performance in two East Coast estuaries. First, the hydrodynamic and transport model was tested

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for the Delaware Bay (MAR). The model was forced with the monthly, minimal and maximal observed discharge at Trenton over the period between 1912 and 1985 (UNH/GRDC Database, Fekete et al., 2000). Simulated salinity profiles are compared with salinity observations from January, February, May and June (the months with the highest number of data entries), which were extracted from the UNH/GRDC Database. Figure 6 shows that the model captures both the salinity intrusion length and the overall shape of the salinity profile well. In addition, the performance of the biogeochemical model and specifically its ability to reproduce pH and pCO_2 profiles was evaluated by a model-data comparison for both the Delaware Bay (MAR) in July 2003 and the Altamaha river estuary (SAR) in October 1995. Similar to Volta et al., 2016a, the test systems were chosen due to their contrasting geometries. The Delaware Bay is a marine dominated system characterized by a pronounced funnel shape, while the Altamaha River has a prismatic estuary characteristic of river dominated systems (Jiang et al., 2008). Monthly upstream boundary conditions for nutrients, as well as observed pH data and calculated pCO_2 are extracted from datasets described in (Sharp, 2010) and (Sharp et al., 2009) for the Delaware and in (Cai and Wang, 1998; Jiang et al., 2008) and (Cai et al., 1998) for the Altamaha river estuary. The additional forcings and boundary conditions are set similarly to the simulation for 2000 (see Tab. 2, 3, 4, 5, 6 in SI). Figure 7 shows that measured and simulated pH values are in good agreement with observed pH and observation-derived calculations of pCO_2. In the Delaware Bay, a pH minimum is located around km 140 and is mainly caused by intense nitrification sustained by large inputs of NH_4 from the Philadelphia urban area, coupled to an intense heterotrophic activity. Both processes lead to a well-developed pCO_2 increase in this area (Fig. 7b). Although no pCO_2 data were available for validation for the period from which boundary conditions were extracted, the simulated profile agree with pCO_2 measurement from July 2013 presented by Joesoef et al. (2015) with pCO_2 values close to equilibrium with the atmosphere in the widest section of the Delaware Bay (close to the estuarine mouth) and values above 1200 µatm at salinities below 5. For the Altamaha river estuary, pH steadily increases from typical river to typical coastal ocean values (Fig. 7b). In addition, both observations and model results reveal that

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outgassing is very intense in the low-salinity region with more than a 5 fold decrease in pCO_2 between salinity 0 and 5 (Fig. 7d).

While such local validations allow assessing the performance of the model for a specific set of conditions, the purpose of this study is to capture the average biogeochemical behavior of the estuaries of the eastern coast of the US. Therefore, in addition to the system-specific validation, published annually averaged FCO_2 estimates for 13 tidal systems located within the study area collected over the 1994-2006 period are compared to simulated FCO_2 for conditions representative of the year 2000. Overall, simulated FCO_2 are comparable to values reported in the literature (Tab. 2). Although discrepancies, which sometimes can significant, are observed at the level of individual systems, the model captures remarkably well the overall trend in CO_2 evasion rate across estuaries. The model simulates low CO_2 efflux (< 5 mol C m^{-2} yr^{-1}) for the 7 systems were such conditions have been observed, while the 6 systems for which the CO_2 evasion exceeds 10 mol C m^{-2} yr^{-1} are the same in the observations and in the model runs. The discrepancy at the individual system level likely result from a combination of factors, including the choice of model processes and there parametrization, the uncertainties in constraining boundary conditions and the limited representability of instantaneous and local observed.

3 Results and discussion

3.1 Spatial variability of estuarine carbon dynamics

Figure 8 presents the spatial distribution of simulated mean annual $\overline{FCO_2}$ and -$\overline{NEM}$ (Fig. 8a), as well as FCO_2 and -*NEM* (Fig. 8b). In general, mean annual $\overline{FCO_2}$ are about 30% larger than mean annual $\overline{NEM}$, with the exception of six estuaries situated in the North of the coastal segment. Overall, the $\overline{NEM}$ is characterized by smaller system to system variability compared to the $\overline{FCO_2}$ in all regions. In addition, Fig. 8 reveals distinct differences across the three coastal segments and highlights the

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important influence of the estuarine geometry and residence time, as well as the latitudinal temperature gradient on estuarine carbon cycling.

Overall, $\overline{FCO_2}$ values are the lowest in the NAR (mean flux = 17.3 ± 16.4 mol C m^{-2} y^{-1}; surface weighted average = 23.1 mol C m^{-2} y^{-1}), consistent with previously reported very low values for small estuaries surrounding the Gulf of Maine (Hunt et al., 2010; 2011; Tab. 2). In contrast, $\overline{NEM}$ reveals a regional minimum in the NAR (-51.2 ± 16.6 mol C m^{-2} y^{-1}; surface weighted average = -52.8 mol C m^{-2} y^{-1}). The MAR is characterized by intermediate values for $\overline{FCO_2}$, with a mean flux of 26.3 ± 34.6 mol C m^{-2} y^{-1} (surface weighted average =11.1 mol C m^{-2} y^{-1}) and lowest values for $\overline{NEM}$ (-15.1 ± 14.2 mol C m^{-2} y^{-1}; surface weighted average =-7.4 mol C m^{-2} y^{-1}). This region also shows the largest variability in CO_2 outgassing compared to the NAR and SAR, with the standard deviation exceeding the mean $\overline{FCO_2}$, and individual estimates ranging from 3.9 mol C m^{-2} y^{-1} to 150.8 mol C m^{-2} y^{-1}. This variability is mainly the result of largely variable estuarine surface areas and volumes. Some of the largest East coast estuaries (e.g. Chesapeake and Delaware Bays), as well as some of smallest estuaries (e.g. York River and Hudson River estuaries, Raymond et al., 1997; 2000), are located in this region (Tab. 2 and 4). The maximum values of 150.8 mol C m^{-2} y^{-1} simulated in the MAR are similar to the highest FCO_2 reported in the literature (132.3 mol C m^{-2} y^{-1} for the Tapti estuary in India; Sarma et al., 2012). The SAR is characterized by the highest mean $\overline{FCO_2}$ (46.7 ± 33.0 mol C m^{-2} y^{-1}; surface weighted average = 40.0 mol C m^{-2} y^{-1}) and intermediate $\overline{NEM}$ (-36.8 ± 24.7 mol C m^{-2} y^{-1}; surface weighted average = -31.2 mol C m^{-2} y^{-1}).

The NAR is characterized by a regional minimum in $\overline{FCO_2}$, and only contributes 4.6% to the total FCO_2 of the East coast of the US, owing to the small cumulative surface area available for gas exchange in its 10 estuarine systems. In contrast, the 18 MAR estuaries, with their large relative contribution to the total regional estuarine surface area, account for as much as 70.1% of the total outgassing. Because of their smaller cumulated surface area compared to those of the MAR, the 14 SAR estuaries account for merely 25.3% of the total outgassing despite their regional maximal $\overline{FCO_2}$.

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A similar, yet slightly less pronounced pattern emerges for the $\overline{NEM}$. The NAR, MAR and SAR respectively contribute 13.7%, 60.7% and 25.6% to the total regional net ecosystem metabolism. The comparatively larger relative contribution of the NAR to the total *NEM* as compared to the total FCO_2 can be explained by the importance of the specific aspect ratio for NEM. A larger ratio of estuarine width b0 and convergence length b corresponds to a more funnel shaped estuary while a low ratio corresponds to a more prismatic geometry (Savenije, 2000; Volta et al., 2014). In the NAR, estuaries are generally characterized by relatively narrow widths and deep-water depths, thus limiting the potential surface area for gas exchange with the atmosphere. However, the relative contribution of each region to the total regional *NEM* and FCO_2 is largely controlled by estuarine surface area. Figure 9 illustrates the cumulative *NEM* (a) and FCO_2 (b) as a function of the cumulative estuarine surface areas. The disproportionate contribution of large estuaries from the MAR translates into a handful of systems (Chesapeake and Delaware Bays and the main tributaries of the former, in particular) contributing to roughly half of the regional *NEM* and FCO_2, in spite of relatively low individual rates per unit surface area. However, the smallest systems (mostly located in the NAR and SAR) nevertheless still contribute a significant fraction to the total regional *NEM* and FCO_2. The 27 smallest systems merely account for less than 10% of the total regional estuarine surface area, yet contribute 38% and 29% to the total regional *NEM* and FCO_2, respectively (Fig. 9). This disproportioned contribution can be mainly attributed to their high individual $\overline{FCO_2}$ and $\overline{NEM}$. This is illustrated by the average simulated $\overline{FCO_2}$ for all 27 smallest systems (calculated as the sum of each estuarine CO_2 outgassing per unit surface area divided by the total number of estuarine systems) which is significantly higher (30.2 mol C m^{-2} y^{-1}) than its surface weighted average (14 mol C m^{-2} y^{-1}). Thereby accounting for the disproportionate contribution of very large systems (calculated as the sum of each estuarine CO_2 outgassing divided by the total estuarine surface area across the region).

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Following the approach used in Regnier et al. (2013), the contribution of each biogeochemical process to *FCO_2* is assessed by evaluating their individual contribution to DIC and ALK changes taking into account the local buffering capacity of an ionic solution when TA and DIC are changing due to internal processes, but ignoring advection and mixing (Zeebe and Wolf-Gladrow 2001). In the present study, we quantify the effect of the NEM on the CO_2 balance, which is almost exclusively controlled by aerobic degradation rates because the contributions of denitrification and NPP to the net ecosystem balance are small. Nitrification, a process triggered by the transport and/or production of NH_4 in oxygenated waters, favors outgassing through its effect on pH, which shifts the acid-base equilibrium of carbonate species and increases the CO_2 concentration. The contribution of supersaturated riverine waters to the overall estuarine CO_2 dynamics is calculated as difference between all the other processes creating or consuming CO_2. Figure 10a presents the contribution of the annually integrated *NEM*, nitrification and evasion of supersaturated, DIC enriched riverine waters to the total outgassing for each system, as well as for individual regions of the domain. The calculation of these annual values is based on the sum of the seasonal fluxes. Model results reveal that, regionally, the *NEM* supports about 50% of the estuarine CO_2 outgassing, while nitrification and riverine DIC inputs sustain about 17% and 33% of the CO_2 emissions, respectively. The relative significance of the three processes described above shows important spatial variability. In the NAR, oversaturated riverine waters and NEM respectively sustain 50% and 44% of the outgassing within the sub-region, while nitrification is of minor importance (6%). In the MAR, the contribution of riverine DIC inputs is significantly lower (~30%) and the main contribution to the outgassing is *NEM* (~50%); nitrification accounting for slightly less than 20% of the outgassing. In the SAR, the riverine contribution is even lower (~20%), and the outgassing is mainly attributed to the *NEM* (~55%) and nitrification (~25%). Therefore, although the model results reveal significant variability across individual systems, a clear latitudinal trend in the contribution to the total *FCO_2* emerge from the analysis; the importance of oversaturated riverine water decreasing from North to South, while *NEM* and nitrification increase along the same latitudinal gradient. The increasing relative importance of

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estuarine biogeochemical processes over riverine DIC inputs as drivers of *FCO_2* along the North-South gradient is largely driven by increasing temperatures from North to South, especially in the SAR region (Tab. SI 1).

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Contrasting patterns across the 3 regions can also be observed with respect to carbon filtering capacities, *CFilt* (Fig. 10b). In the NAR, over 90% of the riverine carbon flux is exported to the coastal ocean. However, in the MAR, the high efficiency of the largest systems in processing organic carbon results in a regional *CFilt* that exceeds 50%. This contrast between the NAR and the MAR and its potential implication for the carbon dynamics of the adjacent continental shelf waters has already been discussed by Laruelle et al. (2015). In the NAR, short estuarine residence results in a much lower removal of riverine carbon by degassing compared to the MAR. Laruelle et al. (2015) suggested that this process could contribute to the weaker continental shelf carbon sink adjacent to the NAR, compared to the MAR. In the SAR, most estuaries remove between 40% and 65% of the carbon inputs. The high temperatures observed and resulting accelerated biogeochemical process rates in this region favor the degradation of organic matter and contribute to increase the estuarine filtering capacity for carbon. However, in the SAR, a large fraction of the OC loads is derived from adjacent salt marshes located along the estuarine salinity gradients, thereby reducing the overall residence time of OC within the systems. The filtering capacity of the riverine OC alone, which transits through the entire estuary, would thus be higher than the one calculated here. As a consequence, highest C retention rates are expected in warm tidal estuaries devoid of salt marshes or mangroves (Cai, 2011).

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3.2 Seasonal variability of estuarine carbon dynamics

Carbon dynamics in estuaries of the US East coast not only show a marked spatial variability, but also vary on the seasonal timescale. Table 5 presents the seasonal distribution of *NEM* and *FCO_2* for each sub-region. In the NAR, a strong seasonality is simulated for the *NEM* and the summer period contributes more than a third to the annually integrated value. The outgassing reveals a lower seasonal variability and is only slightly higher than summer outgassing during fall and lower during spring. In the MAR, summer contributes more to the *NEM* (>28% of the yearly total) than any other season, but seasonality is less pronounced than in the NAR. Here, FCO_2 is largest in winter and particularly low during summer. In the SAR, summer accounts for 30 % of the NEM, while spring contributes 21 %. FCO_2 is relatively constant throughout the year suggesting that seasonal variations in carbon processing decrease towards the lower latitudes in the SAR. This is partly related to the low variability in river discharge throughout the year in lower latitudes (Tab. SI1). In riverine dominated systems with low residence times, such as, for instance, the Altamaha River estuary, the CO_2 exchange at the air-water interface is mainly controlled by the river discharge because the time required to degrade the entire riverine organic matter flux exceeds the transit time of OC through the estuary. Therefore, the riverine sustained outgassing is highest during the spring peak discharge periods. In contrast, the seasonal variability in FCO_2 in long-residence, marine-dominated systems with large marsh areas (e.g. Sapelo and Doboy Sound) is essentially controlled by seasonal temperature variations. Its maximum is reached during summer when marsh plants are dying and decomposing, as opposed to spring when marshes are in their productive stage (Jiang et al., 2008). These contrasting seasonal trends have already been reported for different estuarine systems in Georgia, such as the Altamaha Sound, the Sapelo Sound and the Doboy Sound (Cai, 2011). At the scale of the entire East coast of the US, the seasonal trends in *NEM* reveal a clear maximum in summer and minimal values during autumn and winter. The seasonality of FCO_2 is much less pronounced because the outgassing of oversaturated riverine waters throughout the year contributes to a large fraction of the FCO_2 and dampens the effect of the temperature dependent processes (*NEM* and denitrification). In our simulations, the competition between temperature and river discharge is the main driver of the seasonal estuarine carbon dynamics is. When discharge increases, the carbon loads increase proportionally and the residence time within the system decreases, consequently limiting an efficient degradation of organic carbon input fluxes. In warm

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regions like the SAR, the temperature is sufficiently high all year round to sustain high C processing rates and this explains the reduced seasonal variability in NEM.

3.3 Regional carbon budget: a comparative analysis

The annual carbon budget for the entire East coast of the US is summarized in Fig. 11a. The total carbon input to estuaries along the East coast of the US is 4.6 Tg C y^{-1}, of which 42% arrives in organic form and 58% in inorganic form**.** Of this total input, saltmarshes contribute 0.6 Tg C yr^{-1}, which corresponds to about 14% of the total carbon loads and 32% of the organic loads in the region. The relative contribution of the saltmarshes to the total carbon input increases towards low latitudes and is as high as 60% in the SAR region. Model results suggest that 2.7 Tg C y^{-1} is exported to the continental shelf (25% as TOC and 75% as DIC), while 1.9 Tg C y^{-1} is emitted to the atmosphere. The overall carbon filtering capacity of the region thus equals 41% of the total carbon entering the 43 estuarine systems (river + saltmarshes). Because of the current lack of a benthic module in C-GEM, the water column carbon removal occurs entirely in the form of CO_2 outgassing and does not account for the potential contribution of carbon burial in sediments. The estimated estuarine carbon retention presented here is thus likely a lower bound estimate. Reported to the modeled surface area of the region, the total FCO_2 of 1.9 Tg C y^{-1} translates into a mean air water CO_2 flux of about 14 mol C m^{-2} y^{-1}. This value is slightly higher than the estimate of 10.8 mol C m^{-2} y^{-1} calculated by Laruelle et al., (2013) on the basis of local $\overline{FCO_2}$ estimates assumed to be representative of yearly averaged conditions (see section 2.1). The latter was calculated as the average of 13 annual $\overline{FCO_2}$ values reported in the literature (Tab. 2), irrespective of the size of the systems. This approach is useful and widely used to derive regional and global carbon budgets (Borges et al., 2005; Laruelle et al., 2010; Chen et al., 2013). However, it may lead to potentially significant errors (Volta et al., 2016a) due to the uncertainty introduced by the spatial interpolation

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of local measurements to large regional surface areas, while useful and widely used to derive regional and global carbon budgets.

Regional C budgets are sparse. To our knowledge, the only other published regional assessment of the estuarine carbon and CO_2 dynamics comes from a relatively well studied region: the estuaries flowing into the North Sea in Western Europe (Fig. 11b). This budget was calculated using a similar approach (Volta 2016a) and thus provides an ideal opportunity for a comparative assessment of C cycling in these regions. However, it is important to note that there are also important differences in the applied model approaches and those differences should be taken into account when comparing the derived budgets. In particular, the NW European study is based on a simulation of the 6 largest systems only (Elbe, Scheldt, Thames, Ems, Humber and Weser), accounting for about 40% for the riverine carbon loads of the region. It assumes that the intensity of carbon processing and evasion in all other smaller estuaries discharging into the North Sea (16 % of the carbon loads) can be represented by the average of the 6 largest system simulation results. In addition, the Rhine-Meuse system, which alone accounts for 44% of the carbon riverine inputs of the region, was treated as a passive conduit with respect to carbon due to its very short freshwater residence time (Abril et al., 2002). The contribution of saltmarshes to the regional carbon budget was also ignored because their total surface area is much smaller than along the US East coast (Regnier et al., 2013b). Another important difference is the inclusion of seasonality in the present study while the budget calculated for the North Sea is derived from yearly average conditions (Volta et al., 2016a).

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Overall, although both regions receive similar amounts of C from rivers (4.6 Tg C y^{-1} and 5.9 Tg C y^{-1} for the East coast of the US and the North Sea, respectively), they reveal significantly different C filtering capacities. While the estuaries of the East coast of the US filter 41% of the riverine TC loads, those from the North Sea only remove 8% of the terrestrial-derived material. This is partly due to the large amounts of carbon transiting through the ‘passive’ Rhine-Meuse system. The regional filtering capacity is higher (15%) when this system is excluded from the analysis. However, even when neglecting this system, significant differences in filtering efficiencies between both regions remain. FCO_2 from the North Sea estuaries (0.5 Tg C y^{-1}) is significantly lower than the 1.9 Tg C y^{-1} computed for the East coast of the US. The reason for the lower evasion rate in NW European estuaries is essentially twofold. First, the total cumulative surface area available for gas exchange is significantly lower along the North Sea, in spite of comparable flux densities calculated using the entire estuarine surface areas of both regions (14 mol C m^{-2} y^{-1} and 23 mol C m^{-2} y^{-1} for the East coast of the US and the North Sea, respectively). Second, although the overall riverine carbon loads are comparable in both regions (Fig. 11), the ratio of organic to inorganic matter input is much lower in the North Sea area because of the regional lithology is dominated by carbonate rocks and mixed sediments that contain carbonates (Dürr et al., 2005; Hartmann et al., 2012). As a consequence, TOC represents less than 20% of the riverine loads and only 10% of the carbon exported to the North Sea. In both regions, however, the increase of the inorganic to organic carbon ratio between input and output is sustained by a negative NEM (Fig. 11). Although the ratios themselves may significantly vary from a region of the world to the other as evidenced by these two studies, a NEM driven increase of the inorganic fraction within carbon load along the estuarine axis is consistent with the global estuarine carbon budget proposed by Bauer et al. (2013). In the East coast of the US, the respiration of riverine OC within the estuarine filter is partly compensated by OC inputs from marshes and mangroves in such a way that the input and export IC/OC ratios are closer than in the North Sea region.

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3.4 Scope of applicability and model limitations

Complex multidimensional models are now increasingly applied to quantitatively explore carbon and nutrient dynamics along the land-ocean transition zone over seasonal and even annual timescales (Garnier et al., 2001; Arndt et al., 2007, 2009; Arndt and Regnier, 2007; Mateus et al., 2012). However, the application of such complex models remains limited to individual, well-constrained systems due their high data requirements and computational demand resulting from the need to resolve important physical, biogeochemical and geological processes on relevant temporal and spatial scales. The one-dimensional, computationally efficient model C-GEM has been specifically designed to reduce data requirements and computational demand and to enable regional/global scale applications (Volta et al., 2014, 2016a). However, such a low data demand and computational efficiency inevitably requires simplification. The following paragraphs critically discus these simplifications and their implications.

Spatial resolution

Here, C-GEM is used with a 0.5° spatial resolution. While this resolution captures the features of large systems, it is still very coarse for relatively small watershed, such as those of the St. Francis River, Piscataqua River, May River or the Sapelo River. For instance, the 5 estuaries reported by Hunt et al. (2010, 2011, see section 2.6) are all small systems contained by the same watershed at a 0.5° resolution. Only watersheds whose area spans several grid cells can be properly identified and represented (i.e. Merrimack or Penobscot with 6 and 9 cells, respectively).

Hydrodynamic and Transport Model

C-GEM is based on a theoretical framework that uses idealized geometries and significantly reduces data requirements. These idealized geometries are fully described by three, easily obtainable geometrical parameters (B, b_0, H). The model thus approximates the variability of estuarine width and cross-section along the longitudinal axis through a set of exponential functions. A comprehensive sensitivity study (Volta et al., 2014) has shown that integrated process rates are generally sensitive to changes in these geometrical parameters because of their control on estuarine residence times. For instance, Volta et al. (2014) demonstrated that the NEM, is particularly sensitive to the convergence length. Similarly, the use of constant depth profile may lead to variations of about 10% in NEM (Volta et al., 2014). Nevertheless, geometrical parameters are generally easy to constrain, especially well-monitored regions such as the US east coast. Here, all geometrical parameters are constrained on the basis of observed estuarine surface areas and average water depths. In addition, the model also accounts for the slope of the estuarine channel. This approach ensures that simulated estuarine surface areas, volumes and, thus, residence times are in good agreement with those of the real systems and minimizes uncertainties associated to the physical set-up.

In addition, the one-dimensional representation of the idealized estuarine systems does not resolve two- or three-dimensional circulation features induced by complex topography and density driven circulation. While C-GEM performs well in representing the dominant longitudinal gradients, its applicability to branched systems or those with aspect ratios for which a dominant axis is difficult to identify (e.g. Blackwater estuary, UK; Pearl River estuary, China; Tagus estuary, Portugal; Bay of Brest, France) is limited.

Biogeochemical Model

Although the reaction network of C-GEM accounts for all processes that control estuarine FCO_2 (Borges and Abril, 2012; Cai, 2011), several, potentially important processes, such as benthic-pelagic exchange processes, phosphorous sorption/desorption and mineral precipitation, a more complex representation of the local phytoplankton community, grazing by higher trophic levels, or multiple reactive organic carbon pools are not included. Although these processes are difficult to constrain and their importance for FCO_2 is uncertain, the lack of their explicit representations induces uncertainties in Cfilt. In particular, the exclusion of benthic processes such as organic matter degradation and burial in estuarine sediments could result in an underestimation of Cfilt. However, because very little is known on the long term fate of organic carbon in estuarine sediments, setting up and calibrating a benthic module proves a difficult task. Furthermore, to a certain degree model parameters (such as organic matter degradation and denitrification rate constant) implicitly account for benthic dynamics. We nonetheless acknowledge that, by ignoring benthic processes and burial in particular, our estimates for the estuarine carbon filtering may be underestimated, particularly in the shallow systems of the SAR.

Biogeochemical model parameters for regional and global applications are notoriously difficult to constrain (Volta et al., 2016b). Model parameters implicitly account for processes that are not explicitly resolved and their transferability between systems is thus limited. In addition, published parameter values are generally biased towards temperate regions in industrialized countries (Volta et al., 2016b). A first order estimation of the parameter uncertainty associated to the estuarine carbon removal efficiency (CFilt) can be extrapolated from the extensive parameter sensitivity analyses carried out by Volta et al. (2014, 2016b). These comprehensive sensitivity studies on end-member systems have shown that the relative variation in Cfilt when a number of key biogeochemical parameters are varied by two orders of magnitude varies by is ±15 % in prismatic (short residence time on order of days) to ±25 % in funnel-shaped (long residence time) systems. Thus, assuming that uncertainty increases linearly between those bounds as a function of residence time, an uncertainty estimate can be obtained for each of our modelled estuary. With this simple method, the simulated regional Cfilt of 1.9 Tg C yr-1 would be associated with an uncertainty range comprised between 1.5 and 2.2 Tg C yr^{-1}. Our regional estuarine CO_2 evasion estimate is thus reported with moderate confidence. Furthermore, in the future, this uncertainty range could be further constrained using statistical methods such as Monte Carlo simulations (e.g. Lauerwald et al., 2015).

Boundary Conditions and Forcings

In addition, simulations are only performed for climatological means over the period 1990-2010 without resolving interannual and secular variability. Boundary conditions and forcings are critical as they place the modelled system in its environmental context and drive transient dynamics. However, for regional applications, temporally resolved boundary conditions and forcings are difficult to constrain. C-GEM places the lower boundary condition 20 km from the estuarine mouth into the coastal ocean and the influence of this boundary condition on simulated biogeochemical dynamics is thus limited. At the lower boundary condition, direct observations for nutrients and oxygen are extracted from databases such as the World Ocean Atlas (Antonov et al., 2014). However, lower boundary conditions for OC and pCO_2 (zero concentration for OC and assumption of pCO_2 equilibrium at the sea side) are simplified. This approach does not allow addressing the additional complexity introduced by biogeochemical dynamics in the estuarine plume (see Arndt et al., 2011). Yet, these dynamics only play a secondary role in the presented study that focuses on the role of the estuarine transition zone in processing terrestrial-derived carbon.

Constraining upper boundary conditions and forcings is thus more critical. Here, C-GEM is forced by seasonally-averaged conditions for Q, T, and radiation. To date, GlobalNEWS only provide yearly-averaged conditions for a number of upper boundary conditions (Seitzinger et al., 2005; Mayorga et al., 2010), representative of the year 2000. Simulations are thus only partly transient (induced by seasonality in Q, T and radiation) and do not resolve short-lived events such as storms or extreme drought conditions. In addition, direct observations of upper boundary conditions are rarely available- in particular over seasonal or annual timescales. For the US East Coast estuaries, direct observations are only available for O_2, chlorophyll a, DIC and Alk. For DIC and alkalinity and boundary conditions are constrained by calculating the average concentration over a period of about three decades. In addition, observational data is extracted at the station closest to the model's upper boundary, which might be still located several kilometres upstream or downstream of the model boundary. Upper boundary conditions of POC, DOC, DIN, DIP, DSi are extracted from GlobalNews and thus model-derived. As a consequence, our results are thus intimately dependent on the robustness of the GlobalNEWS predictions. These values are usually only considered robust estimates for watersheds larger than ~10 cells (Beusen et al., 2005), which only correspond to 13 of the 43 estuaries modelled in this study.

Model-data comparison

The generic nature of the applied model approach and, in particular the application of seasonally/annually averaged or model-deduced boundary conditions renders a direct validation of model results on the basis of local and instantaneous observational data (e.g. longitudinal profiles), which is likely not representative of these long-term average conditions, difficult. Therefore, model performance is evaluated on the basis of spatially aggregated estimates (e.g. regional FCO_2 estimates based on local measurements) rather than system-to-system comparisons with longitudinal profile from specific days. However, note that the performance of C-GEM has been intensively tested by specific model-data comparisons for a number of different systems (e.g. Volta et al., 2014, 2016a) and we are thus confident of its predictive capabilities.

Despite the numerous simplifying assumptions inevitably required for such a regional assessment of carbon fluxes along the land-ocean continuum, the presented approach does nevertheless provide an important step forward in evaluating the role of land-ocean transition systems in the global carbon cycle. It provides a first robust estimate of carbon dynamics based on a theoretically well-founded and carefully tested, spatially and temporally resolved model approach. This approach provides novel insights that go beyond those gained through traditionally applied zero-salinity method or box model approaches. In addition, it also highlights critical variables and data gaps and thus helps guide efficient monitoring strategies.

3.5 Towards predictors of the estuarine carbon processing

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The mutual dependence between geometry and transport in tidal estuaries and, ultimately, their biogeochemical functioning (Savenije, 1992; Volta et al., 2014) allows relating easily extractable parameters linked to their shape or their hydraulic properties to biogeochemical indicators. In this section, we explore the relationships between such simple physical parameters and indicators of the estuarine carbon processing $\overline{NEM}$, $\overline{FCO_2}$ and *CFilt*. In order to account for the effect of temperature on C dynamics, -$\overline{NEM}$ and $\overline{FCO_2}$ are also normalized to the same temperature (arbitrarily chosen to be 0 degree). These normalized values are obtained by dividing -$\overline{NEM}$ and $\overline{FCO_2}$ by a Q_{10} function *f(T)* (see Volta et al., 2014). This procedure allows accounting for the exponential increase in the rate of several temperature dependent processes contributing to the NEM (i.e. photosynthesis, organic

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carbon degradation…). Applying the same normalization to -$\overline{NEM}$ and $\overline{FCO_2}$ is a way of testing how intimately linked *NEM* and *FCO_2* are in estuarine systems. Indeed linear relationships relating one to the other have been reported (Mayer and Eyre, 2012). The three indicators are then investigated as a function of the ratio between the estuarine surface *S* and the seasonal river discharge *Q*. The surface area is calculated from the estuarine width and length, as described by equation 2, in order to use a parameter which is potentially applicable to other regions for which direct estimates of the real estuarine surface area is not available. Since the fresh water residence time of a system is obtained by dividing volume by river discharge, the S/Q ratio is also intimately linked to residence time. Here, we choose to exclude the estuarine depth from the analysis because this variable cannot be easily quantified from maps or remote sensing images and would thus compromise the applicability of a predictive relationship on the global scale. However, from dimensional analysis, S/Q can be viewed as a water residence time normalized to meter depth of water. As shown by equation 3, *S* only requires constraining *B0* and width convergence length *b*, two parameters that can readily be extracted from the Google Earth engine. Global database of river discharges, as for instance RivDIS (Vörösmarty et al., 1996) are also available in such a way that the S/Q ratio can potentially be extracted for all estuaries around the globe.

Figure 12a reveals that small values of S/Q are associated with the most negative $\overline{NEM}$ / $f(T)$. The magnitude of the $\overline{NEM}$ then exponentially decreases with increasing values of S/Q. Estuaries characterized by small values of *S/Q* are mainly located in the NAR sub-region and correspond to small surface area, and thus short residence time systems. It is possible to quantitatively relate -$\overline{NEM}$ $/f(T)$ and S/Q through a power law function (y = 25.85 $x^{-0.64}$ with a r^2 = 0.82). The coefficient of determination remains the same when excluding estuaries from the NAR region and the equation itself is not significantly different, although those estuaries on their own do not display any statistically significant trend (Tab. 6). The decrease in the intensity of the net ecosystem metabolism in larger estuaries (Fig 8), characterized by high S/Q ratios, can be related to the extensive

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consumption of the organic matter pool during its transit through the estuarine filter. However, when reported to the entire surface area of the estuary, larger systems (with high values of S/Q) still reveal the most negative surface integrated *NEM* (Fig. 12b). It can also be noted that some estuaries from the NAR region display very low values of –*NEM*. These data points correspond to fall and winter simulations for which the temperature was relatively cold (<5 °C) and biogeochemical processing was very low.

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The overall response of $\overline{FCO_2}/f(T)$ to S/Q is comparable to that of -$\overline{NEM}/f(T)$ (Fig. 12c), with lower values of $\overline{FCO_2}$ observed for high values of S/Q. However, for S/Q < 3 days m^{-1}, the $\overline{FCO_2}$ values are very heterogeneous and contain many, low $\overline{FCO_2}$ outliers from the NAR region. These data points generally correspond to low water temperature conditions which keep pCO_2 low, even if the system generates enough CO_2 internally via NEM. Thus, the well-documented correlation between $\overline{NEM}$ and $\overline{FCO_2}$ (Maher and Eyre, 2012) does not seem to hold for systems with very short residence times. For systems with S/Q > 3 days m^{-1}, we obtain a regression FCO_2 = -0.64 x *NEM* + 5.96 with a r^2 of 0.46, which compares well with the relation FCO_2 = -0.42 x *NEM* + 12 proposed by Maher and Eyre (2012) who used 24 seasonal estimates from small Australian estuaries. However, our results suggest that this relationship cannot be extrapolated to small systems such as those located in the NAR. Figure 12d, which reports non-normalized FCO_2 reveals a monotonous increase of FCO_2 with S/Q. This suggests that, unlike the *NEM* for which the normalization by a temperature function allowed explaining most of the variability; FCO_2 is mostly controlled by the water residence time within the system. Discharge is the main FCO_2 driver in riverine dominated systems, while interactions with marshes are driving the outgassing in marine dominated systems surrounded by marshes. Net aquatic biological production (NEM being negative or near 0) in large estuaries (with large S/Q) is another important reason for low FCO_2 in such systems. For example, despite the higher CO_2 degassing flux in the upper estuary of the Delaware, strong biological CO_2 uptake in the mid-bay and near zero NEM in the lower bay result in a much lower FCO_2 for the entire estuary (Joesoef et al.

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2015). In systems with S/Q < 3 days m^{-1}, the short residence time prevents the excess CO_2 of oversaturated water from being entirely exchanged with the atmosphere and simulations reveal that the estuarine waters are still oversaturated in CO_2 at the estuarine mouth. Thus, the inorganic carbon, produced by the decomposition of organic matter, is not outgassed within the estuary but exported to the adjacent continental shelf waters. This result is consistent with the observation-based hypothesis of Laruelle et al. (2015) for the NAR estuaries. As a consequence of the distinct behavior of short residence time systems, the coefficient of determination of the best-fitted power law function relating $\overline{FCO_2}$ and S/Q is only significant if NAR systems are excluded ($y = 31.64\ x^{-0.58}$ with a $r^2 = 0.70$). This thus suggest that such relationships (as well as that proposed by Maher and Eyre, 2012) cannot be applied to any system but only those for which S/Q>3 day m^{-1}.

Finally, Fig. 12e reports the simulated mean seasonal carbon filtering capacities as a function of the depth normalized residence time. Not surprisingly, and in overall agreement with previous studies on nutrient dynamics in estuaries (Nixon et al., 1996), the carbon filtering capacity increases with S/Q. The best statistical relation between *CFilt* and S/Q is obtained when including all 3 regions, resulting in $r^2 = 0.70$ ($y = 40.64 \log_{10}(x) + 11.84$). Very little C removal occurs in systems with S/Q < 1 day m^{-1}. For systems characterized by longer depth-normalized residence times, *CFilt* increases regularly, and reaches 100% for S/Q > 100 day m^{-1}. Such high values are only observed for very large estuaries from the MAR region (Delaware and Chesapeake Bays); the majority of our systems had an *S/Q* range between 1 and 100 day m^{-1}. The quantitative assessment of estuarine filtering capacities is further complicated by the complex interplay of estuarine and coastal processes. Episodically, marked spatial variability in concentration gradients near the estuarine mouth may lead to a reversal of net material fluxes from coastal waters into the estuary (Regnier at al., 1998; Arndt et al. 2011). Our results show that this feature is particularly significant for estuaries with a large width at the mouth and short convergence length (funnel shaped or 'Bay type' systems). These coastal nutrient and carbon inputs influence the internal estuarine C dynamics and lead to filtering capacities that

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can exceed 100%. This feature is particularly significant in summer, when riverine inputs are low and the marine material is intensively processed inside the estuary.

Previous work investigated the relationship between fresh water residence time and nutrient retention (Nixon et al., 1996; Arndt et al., 2011; Laruelle, 2009). These studies, however, were constrained by the scarcity of data. For instance, the pioneering work of Nixon et al. (1996) only relied on a very limited number (<10) of quite heterogeneous coastal systems, all located along the North Atlantic. Here, our modeling approach allows us to generate 172 (43 x 4) data points, each representing a system-scale biogeochemical behavior. Together, this database spans the entire spectrum of estuarine settings and climatic conditions found along the East coast of the US. In addition, the ratio S/Q used as master variable for predicting temperature normalized -$\overline{NEM}$, $\overline{FCO_2}$ and *CFilt* only requires a few easily accessible geometric parameters (B0, b and L) and an estimate of the river discharge. While it is difficult to accurately predict $\overline{FCO_2}$ for small systems such as those located in the NAR region, the relationships found are quite robust for systems in which S/Q > 3 days m^{-1}. Most interestingly, *CFilt* values reveal a significant correlation with S/Q and could be used in combination with global riverine carbon delivery estimates such as GlobalNews 2 (Mayorga et al., 2010) to constrain the estuarine CO_2 evasion and the carbon export to the coastal ocean at the continental and global scales.

4. Conclusions

This study presents the first complete estuarine carbon budget for the East coast of the US using a modeling approach. The structure of the model C-GEM relies on a restricted number of readily available global datasets to constrain boundary conditions and limits the number of geometrical and physical parameters to be constrained. Our simulations predict a total CO_2 outgassing of 1.9 Tg C y^{-1} for all tidal estuaries of the East coast of the US. This quantification accounts for the seasonality in estuarine carbon processing as well as for distinct individual behaviors among estuarine types (marine or river dominated). The total carbon output to the coastal ocean is estimated at 2.7 TgC y^{-1}, and the carbon filtering capacity with respect to riverine, marshes and mangrove inputs is thus on the order of 40%. This value is significantly higher than the recently estimated C filtering capacity for estuaries surrounding the North Sea using a similar approach (Volta et al., 2016a), mainly because the surface area available for gas exchange and the draining lithology limits the CO_2 evasion in the NW European systems. At the regional scale of the US East coast estuaries, net heterotrophy is the main driver (50%) of the CO_2 outgassing, followed by the ventilation of riverine supersaturated waters entering the estuarine systems (32%) and nitrification (18%). The dominant mechanisms for the gas exchange and the resulting carbon filtering capacities nevertheless reveal a clear latitudinal pattern, which reflects the shapes of estuarine systems, climatic conditions and dominant land-use characteristics.

Our model results are used to derive predictive relationships relating the intensity of the area-based Net Ecosystem Metabolism ($\overline{NEM}$), air-water CO_2 exchange ($\overline{FCO_2}$) and the carbon filtering capacity (*CFilt*) to the depth normalized residence time, expressed as the ratio of the estuarine surface area to the river discharge. In the future, such simple relationships relying on readily available geometric and hydraulic parameters could be used to quantify carbon processing in areas of the world devoid of direct measurements. However, it is important to note that such simple relationships are only valid over the range of boundary conditions and forcings explored and may not be applicable to conditions that fall outside of this range. In regions with better data coverage, such as the one investigated here, our study highlights that the regional-scale quantification, attribution, and projection of estuarine biogeochemical cycling are now at reach.

5. Acknowledgements

G. G. Laruelle is Chargé de recherches du F.R.S.-FNRS at the Université Libre de Bruxelles. The research leading to these results has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 643052

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(C-CASCADES project). The authors thank V. L. Mulder for her thorough reading of the manuscript upon submission.

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Table 1: Estimates of total annual riverine input from watersheds to estuaries (Tg C yr^{-1}). The ranges are based on Stets and Striegl (2012), Global NEWS (Mayorga et al. 2010), Hartmann et al. (2009), SPARROW (Shih et al. 2010) and DLEM (Tian et al. 2010, 2012). Modified from Najjar et al. 2012.

	DIC	**DOC**	**POC**	**TOTAL**
NAR	0.2-0.8	0.3-2.1	0.1-0.2	0.6-3.1
MAR	1.4-1.8	0.5-2.3	0.1-0.3	2.0-4.4
SAR	0.4-1.4	0.9-1.6	0.1-0.2	1.4-3.2
TOTAL	2.0-4.0	1.7-6.0	0.3-0.7	4.0-10.7

Table 2: Published local annually averaged estimates of $\overline{FCO_2}$ in mol C m^{-2} yr^{-1} for estuaries along the East coast of the US."

Name	**Lon**	**Lat**	$\overline{FCO_2}$ **Observed.**	**Modeled**	**Reference**
Altamaha Sound	-81.3	31.3	32.4	72.7	Jiang et al. (2008)
Bellamy	-70.9	43.2	3.6	3.9	Hunt et al. (2010)
Cocheco	-70.9	43.2	3.1	3.9	Hunt et al. (2010)
Doboy Sound	-81.3	31.4	13.9	25.7	Jiang et al. (2008)
Great Bay	-70.9	43.1	3.6	3.9	Hunt et al. (2011)
Little Bay	-70.9	43.1	2.4	3.9	Hunt et al. (2011)
Oyster Bay	-70.9	43.1	4	3.9	Hunt et al. (2011)
Parker River estuary	-70.8	42.8	1.1	3.9	Raymond and Hopkinson (2003)
Sapelo Sound	-81.3	31.6	13.5	20.6	Jiang et al. (2008)
Satilla River	-81.5	31	42.5	25.7	Cai and Wang (1998)
York River	-76.4	37.2	6.2	8.1	Raymond et al. (2000)
Hudson River	-74	40.6	13.5	15.5	Raymond et al. (1997)
Florida Bay	-80.68	24.96	1.4	n.a.	Dufore (2012)

Table 3: State variables and processes explicitly implemented in CGEM.

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State variables		
Name	**Symbol**	**Unit**
Suspended Particulate Mater	SPM	gL^{-1}
Total Organic Carbon	TOC	µM C
Nitrate	NO_3	µM N
Ammonium	NH_4	µM N
Phosphate	DIP	µM P
Dissolved Oxygen	DO	µM O_2
Phytoplankton	Phy	µM C
Dissolved Silica	dSi	µM Si
Dissolved Inorganic Carbon	DIC	µM C
Biogeochemical reactions		
Name	**Symbol**	**Unit**
Gross primary production	GPP	µM C s^{-1}
Net primary production	NPP	µM C s^{-1}
Phytoplankton mortality	M	µM C s^{-1}
Aerobic degradation	R	µM C s^{-1}
Denitrification	D	µM C s^{-1}
Nitrification	N	µM N s^{-1}
O_2 exchange with the atmosphere	FO_2	µM O_2 s^{-1}
CO_2 exchange with the atmosphere	FCO_2	µM C s^{-1}
SPM erosion	E_{SPM}	gL^{-1} s^{-1}
SPM deposition	D_{SPM}	gL^{-1} s^{-1}

Table 4: Yearly averaged surface area (*S*), fresh water discharge (*Q*), residence time (*Rt*), FCO_2 and *NEM* of all simulated estuaries.

long degrees	lat degrees	*S* km^2	*Q* $m^3 s^{-1}$	*Rt* days	$\overline{FCO_2}$ mol C m^{-2} yr^{-1}	$\overline{NEM}$ mol C m^{-2} yr^{-1}	FCO_2 10^6 mol C yr^{-1}	*NEM* 10^6 mol C yr^{-1}
NAR								
-67.25	44.75	7	38.5	15	3.7	-37.4	27	-270
-67.25	45.25	12	73.6	15	6.0	-56.7	71	-666
-67.25	45.25	12	73.6	15	13.8	-56.6	162	-666
-67.75	44.75	3	68.5	4	6.7	-63.5	23	-221
-68.25	44.75	14	69.5	19	4.1	-56.2	58	-791
-68.75	44.75	89	309.9	23	27.4	-58.2	2431	-5163
-69.75	44.25	50	626.6	5	32.3	-74.4	1607	-3703
-70.25	43.75	3	25.8	10	2.1	-21.0	7	-71
-70.75	41.75	288	103.6	958	5.0	-4.0	1428	-1146
-70.75	42.25	63	210.7	40	16.2	-32.9	1025	-2081
-70.75	42.75	17	105.8	3	56.3	-69.0	943	-1155
MAR								
-70.75	43.25	31	29.9	11	21.6	-37.4	662	-1146
-71.25	41.75	257	28.2	808	3.9	-2.5	997	-650
-71.75	41.25	21	112.4	4	35.2	-32.6	726	-672
-72.75	40.75	20	25.4	62	30.7	-21.1	623	-430
-72.75	41.25	10	142.5	2	150.8	-36.9	1578	-386
-72.75	41.75	55	476.6	3	55.9	-45.7	3088	-2523
-73.25	40.75	19	26.8	56	31.4	-28.4	608	-550
-74.25	40.75	1192	608.2	126	15.5	-11.8	18432	-14047
-75.25	38.25	399	80.5	172	13.9	-5.0	5558	-2016
-75.25	38.75	354	31.8	357	7.5	-3.0	2659	-1076
-75.25	39.75	1716	499.0	221	10.0	-7.8	17072	-13439
-75.75	39.25	224	18.3	434	7.5	-2.9	1685	-640
-76.25	39.25	3427	717.1	352	8.1	-5.1	27646	-17352
-76.75	37.25	586	272.3	74	15.0	-10.4	8810	-6084
-76.75	37.75	154	36.3	163	10.7	-6.6	1654	-1023
-76.75	39.25	59	71.2	29	48.6	-34.6	2862	-2038
-77.25	38.25	206	30.2	268	6.1	-3.3	1265	-676
-77.25	38.75	568	259.2	118	16.7	-10.8	9488	-6134
SAR								
-78.25	34.25	48	167.4	7	122.5	-62.4	5916	-3015
-79.25	33.25	47	56.3	42	43.4	-36.5	2056	-1728
-79.25	33.75	45	291.4	8	85.1	-78.7	3843	-3551
-79.75	33.25	25	33.8	15	37.9	-32.8	956	-828
-80.25	32.75	25	31.0	50	48.8	-42.5	1214	-1057
-80.25	33.25	92	75.5	61	62.7	-61.2	5769	-5625
-80.75	32.25	71	21.1	182	12.9	-7.0	918	-501
-80.75	32.75	164	63.1	95	20.6	-11.5	3372	-1879
-81.25	31.75	92	71.7	45	25.7	-20.9	2361	-1926
-81.25	32.25	130	379.8	11	51.7	-39.2	6732	-5097
-81.75	30.75	34	18.7	61	17.5	-14.7	602	-505
-81.75	31.25	130	17.7	294	5.5	-4.0	713	-523
-81.75	31.75	56	350.5	4	72.7	-67.4	4068	-3770

Deleted: **Table 3**: Published local annually averaged estimates of $\overline{FCO_2}$ for estuaries along the East coast of the US. ¶
Name

Table 5: Seasonal contribution to FCO_2 and *NEM* in each the sub-region. The seasons displaying the highest percentages are indicated in bold. Winter is defined as January, February and March, Spring as April, May and June and so on…

Region	*NEM* mol C y^{-1}	winter %	spring %	summer %	fall %	FCO_2 mol C y^{-1}	winter %	spring %	summer %	fall %
NAR	$-16.3\ 10^9$	14.7	21.2	**37.0**	27.2	$7.2\ 10^9$	26.3	18.9	26.5	**28.3**
MAR	$-72.2\ 10^9$	21.9	25.9	**28.8**	23.4	$108.3\ 10^9$	**29.8**	23.3	20.7	26.2
SAR	$-30.5\ 10^9$	24.6	20.9	**30.3**	24.2	$39.2\ 10^9$	26	23.4	**27**	23.6

Table 6: Regressions and associated coefficient of determination between the depth normalized residence time (S/Q) and -$\overline{NEM}$ / *f(T)*, $\overline{FCO_2}$ / *f(T)* and *CFilt*.

Region	$-\overline{NEM}/f(T)$	$\overline{FCO_2}/f(T)$	*CFilt*
NAR	$y = 27.84\ x^{-0.17}$ $r^2 = 0.11$	$y = 6.07\ x^{0.00}$ $r^2 = 0.00$	$y = 15.08 \log_{10}(x) + 4.86$ $r^2 = 0.40$
MAR	$y = 26.03\ x^{-0.63}$ $r^2 = 0.86$	$y = 34.36\ x^{-0.58}$ $r^2 = 0.68$	$y = 40.46 \log_{10}(x) + 9.60$ $r^2 = 0.70$
SAR	$y = 28.36\ x^{-0.71}$ $r^2 = 0.76$	$y = 32.82\ x^{-0.66}$ $r^2 = 0.80$	$y = 23.19 \log_{10}(x) + 43.71$ $r^2 = 0.46$
MAR + SAR	$y = 25.85\ x^{-0.64}$ $r^2 = 0.82$	$y = 31.64\ x^{-0.58}$ $r^2 = 0.70$	$y = 33.30 \log_{10}(x) + 24.88$ $r^2 = 0.57$
NAR + MAR + SAR	$y = 28.98\ x^{-0.66}$ $r^2 = 0.82$	$y = 12.98\ x^{-0.33}$ $r^2 = 0.30$	$y = 40.64 \log_{10}(x) + 11.84$ $r^2 = 0.70$

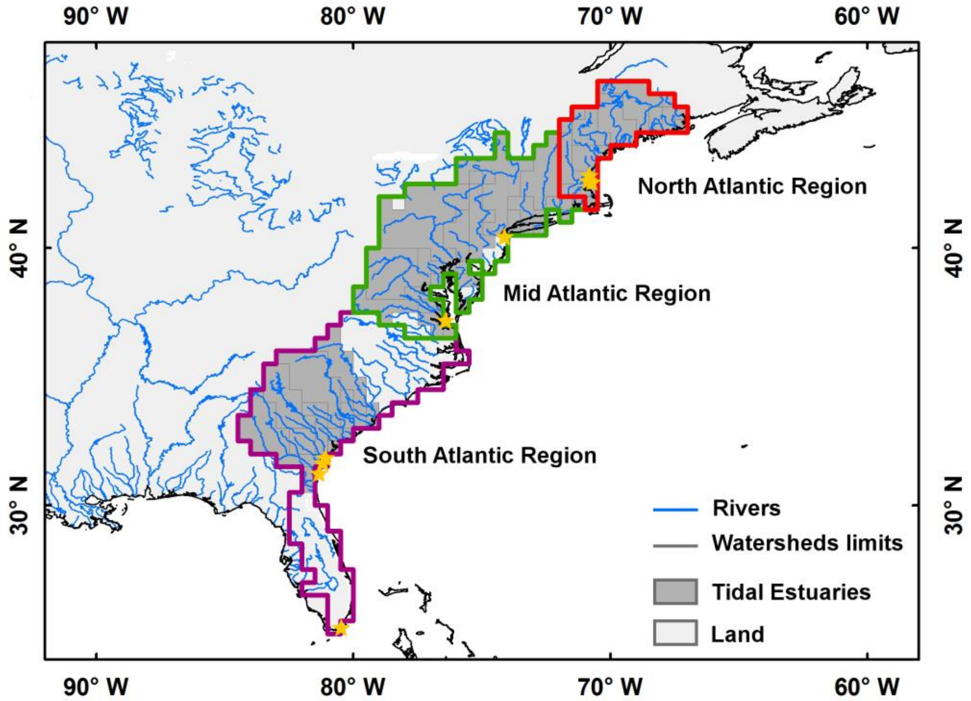
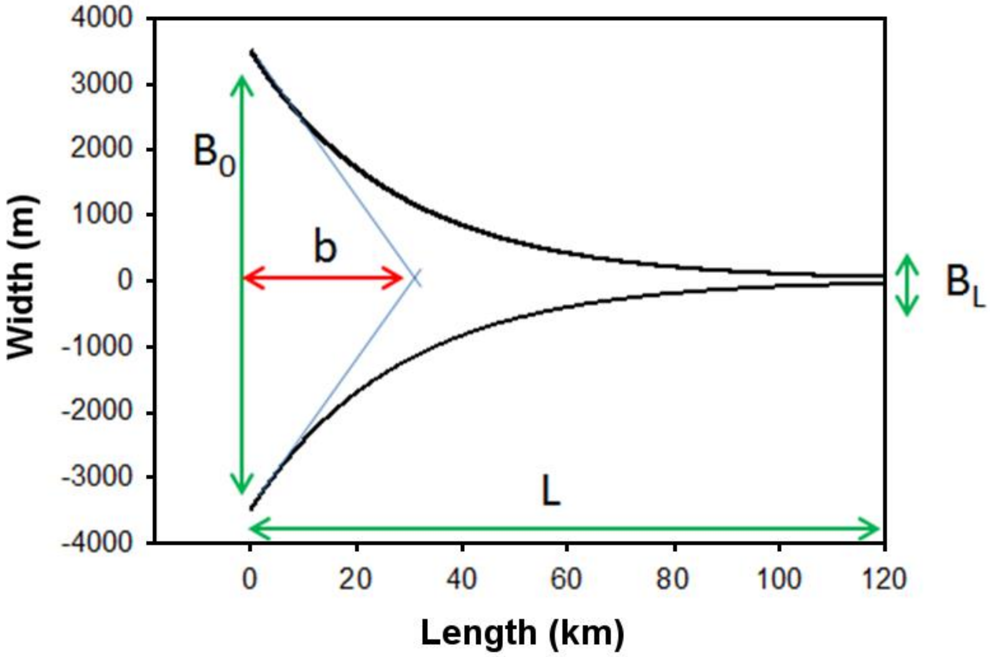


Figure 1: Limits of the 0.5 degrees resolution watersheds corresponding to tidal estuaries of the East coast of the US. 3 sub-regions are delimited with colors and orange stars represent the location of previous studies.



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1747 **Figure 2:** Idealized estuarine geometry and main parameters. Parameters indicated by green arrows
1748 are measured, b is calculated. See section 2.3.1 for further details.

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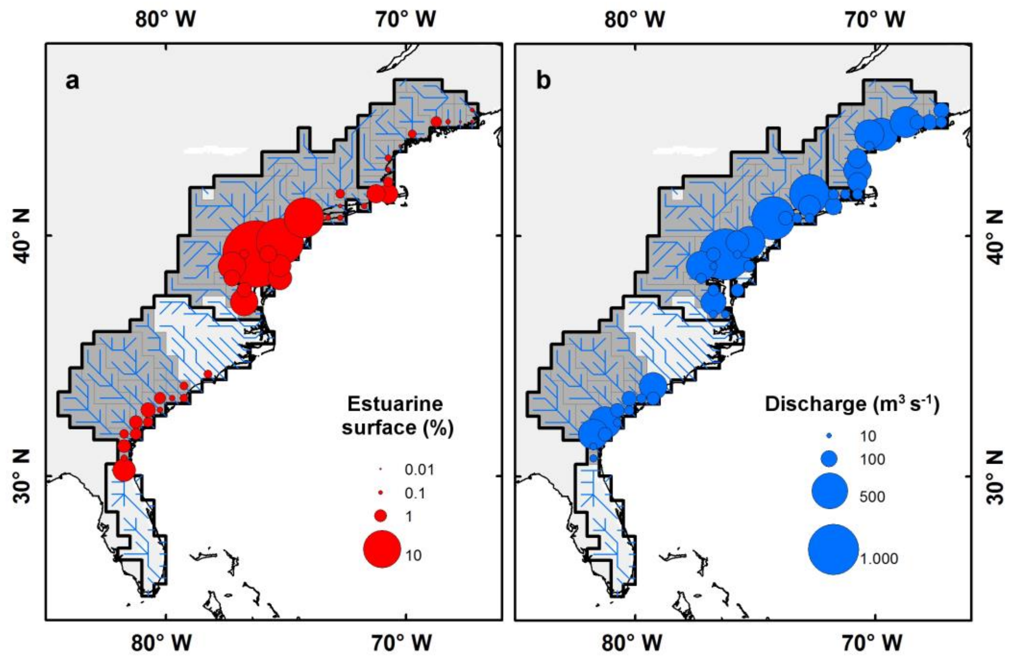

Figure 3: Estuarine surface area (a) and mean annual freshwater discharge (b) for each tidal estuary of the East coast of the US. Estuarine surface area are expressed as percentage of the entire surface area of the region (19830 km^2)

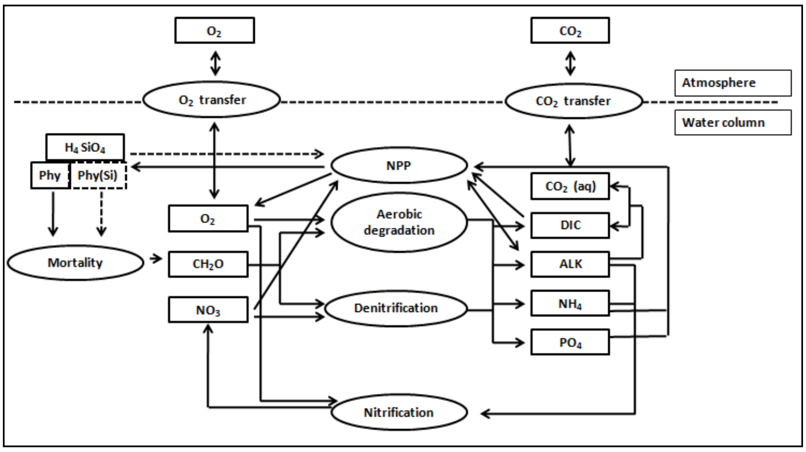

Figure 4: Conceptual scheme of the biogeochemical module of C-GEM used in this study. State-variables and processes are represented by boxes and oval shapes, respectively. Modified from Volta et al., 2014.

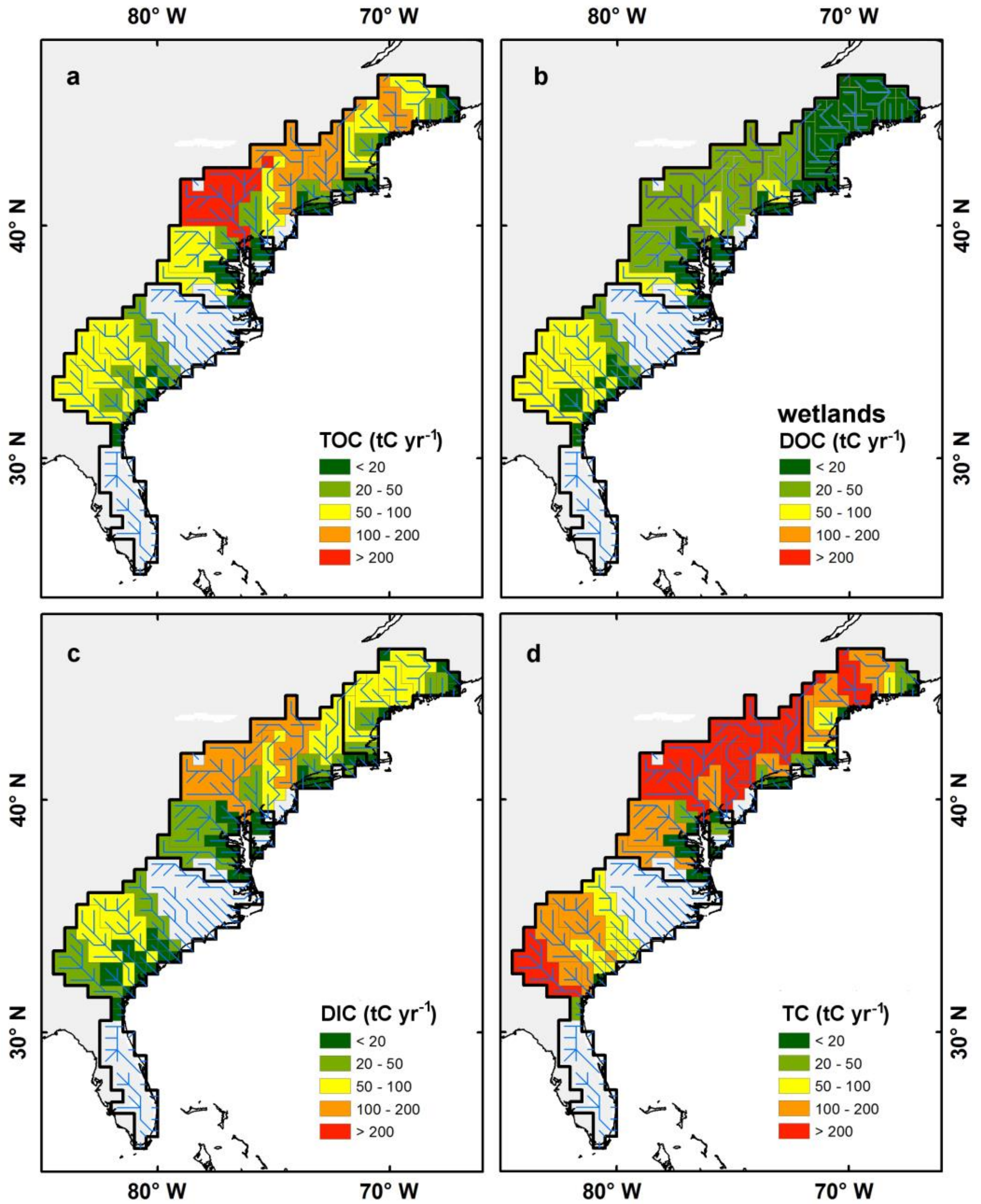

Figure 5: Annual river carbon loads of TOC (a), annual DOC fluxes from wetlands (b), annual river carbon loads of DIC (c) and annual TC fluxes (d). All fluxes are indicated per watershed.

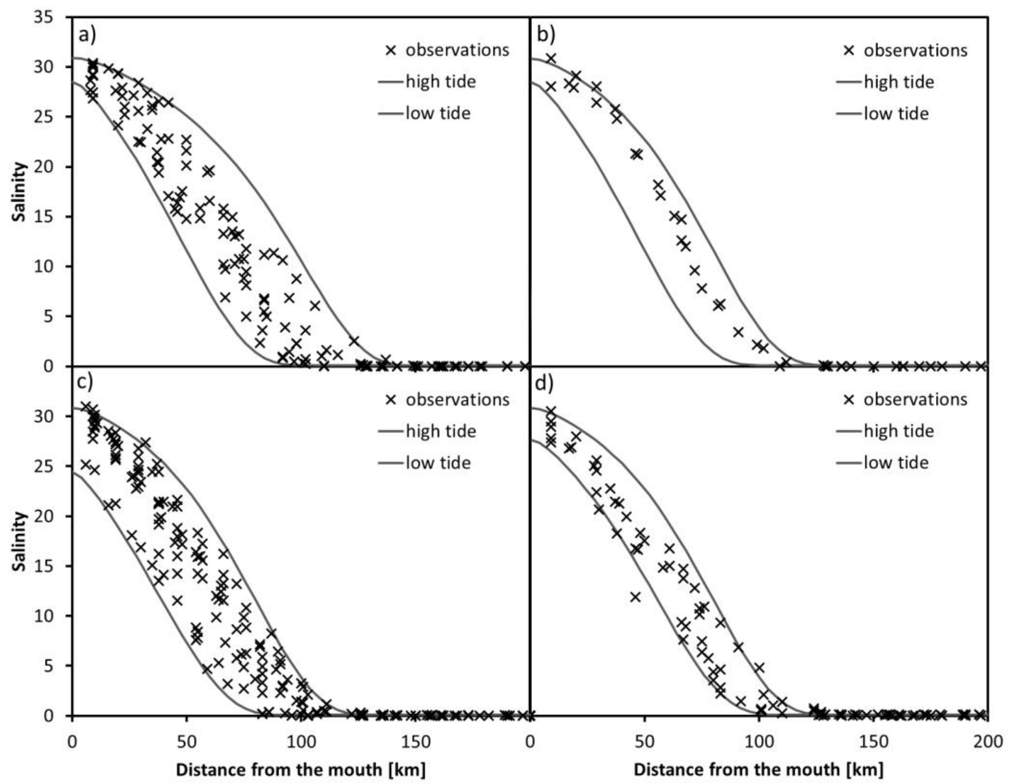

Figure 6. Modeled (lines) and measured (crosses) salinities in the Delaware Bay estuary for January (a), February (b), May (c), June (d). The two lines correspond to high and low tides.

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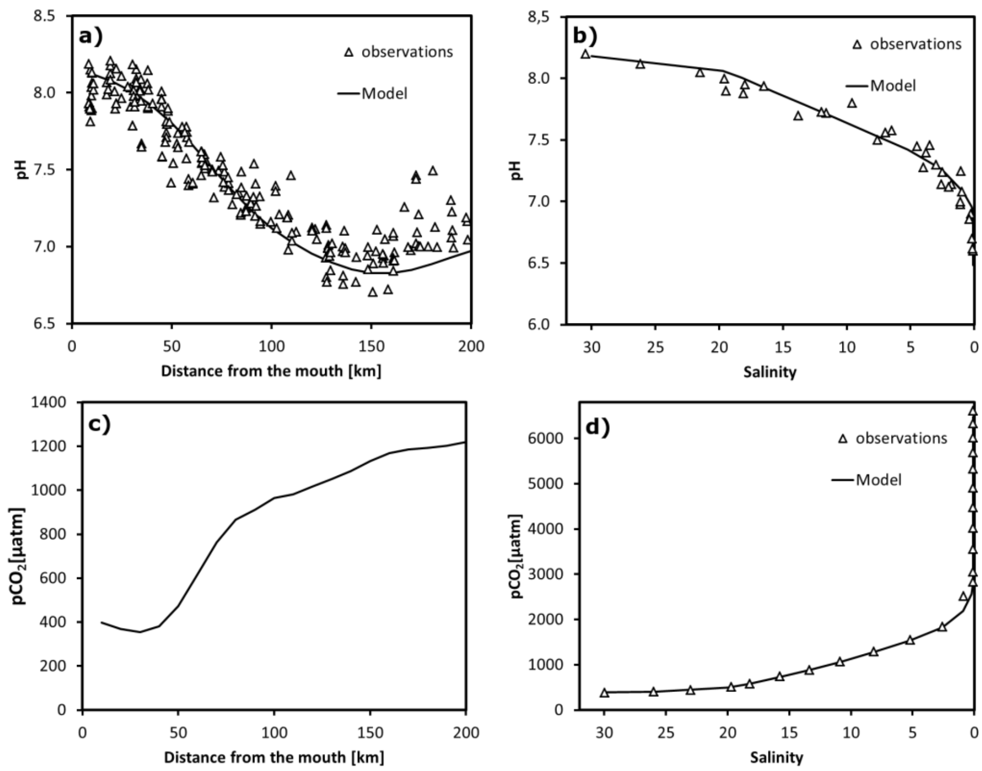

Figure 7. Longitudinal profiles of pH (top) and pCO_2 (bottom) for the Delaware Bay (left) and Altamaha river estuary (right).

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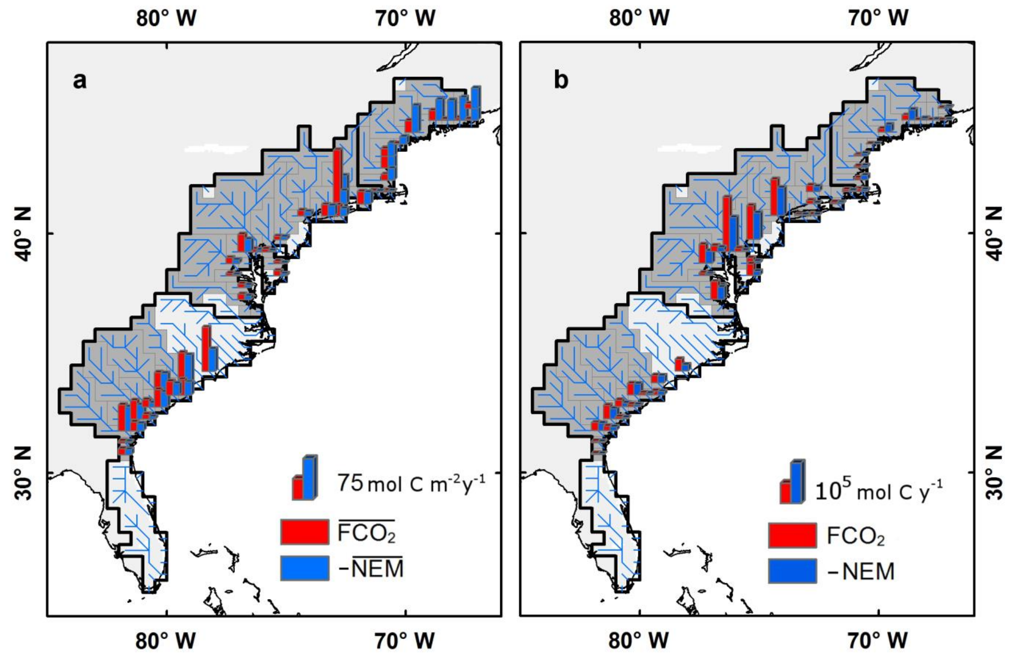

Figure 8: Spatial distribution of spatially averaged value (a) and integrated value (b) of mean annual *FCO_2* (red) and *-NEM* (blue) along the East coast of the US. On panel a, the notation with overbars ($\overline{FCO_2}$ and -$\overline{NEM}$) represents rates per unit surface. For the sake of the comparison with $\overline{FCO_2}$, Fig. 8 displays -$\overline{NEM}$ because the model predicts that all estuaries in this region are net heterotrophic.

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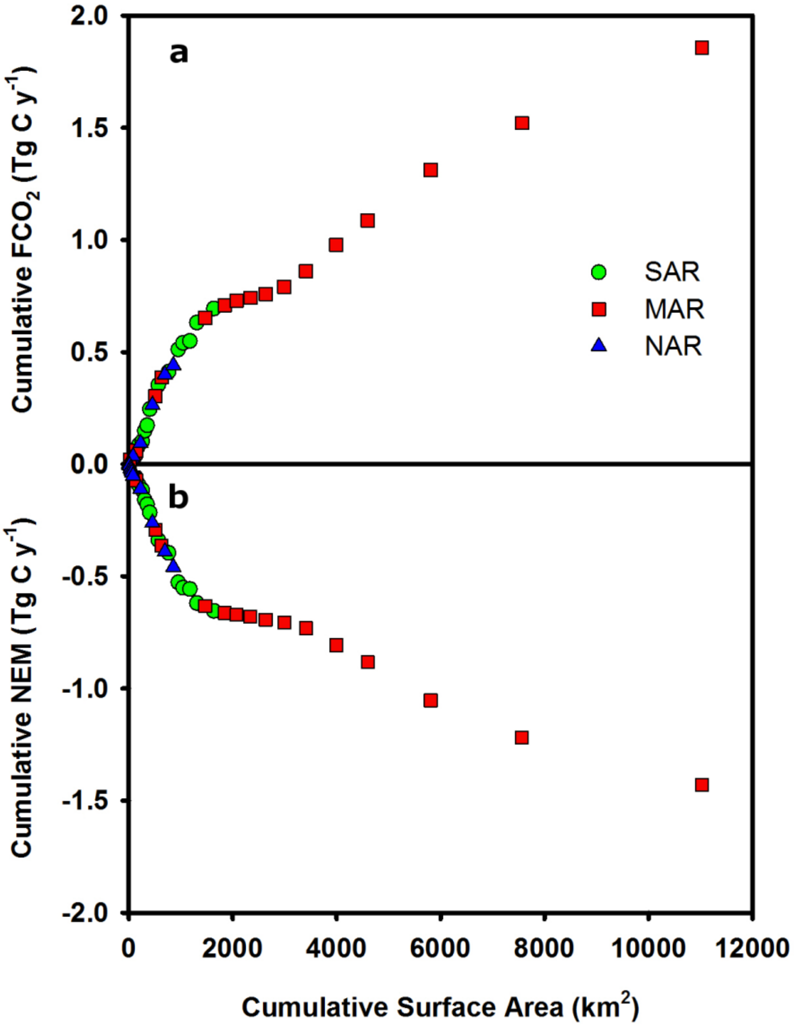

Figure 9: The Cumulative FCO_2 (a) and *NEM* (b) as functions of the cumulative estuarine surface area. Systems are sorted by increasing surface area.

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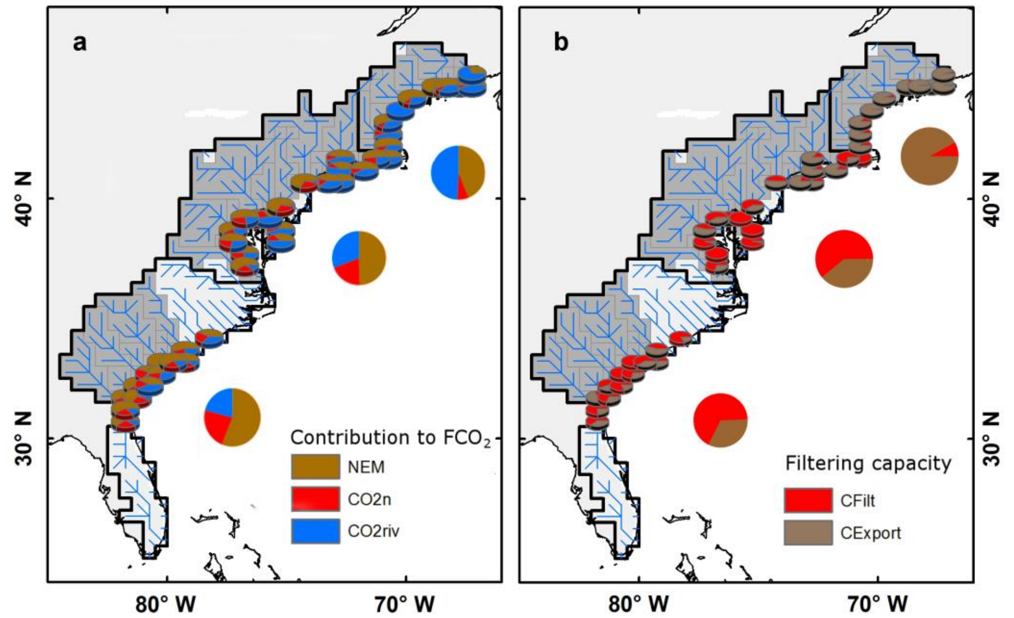

Figure 10: Contribution of *NEM*, nitrification and riverine waters super-saturated waters to the mean annual FCO_2 (a). Spatial distribution of mean annual carbon filtration capacities (*CFilt*) and export (*CExport*) along the East coast of the US (b).

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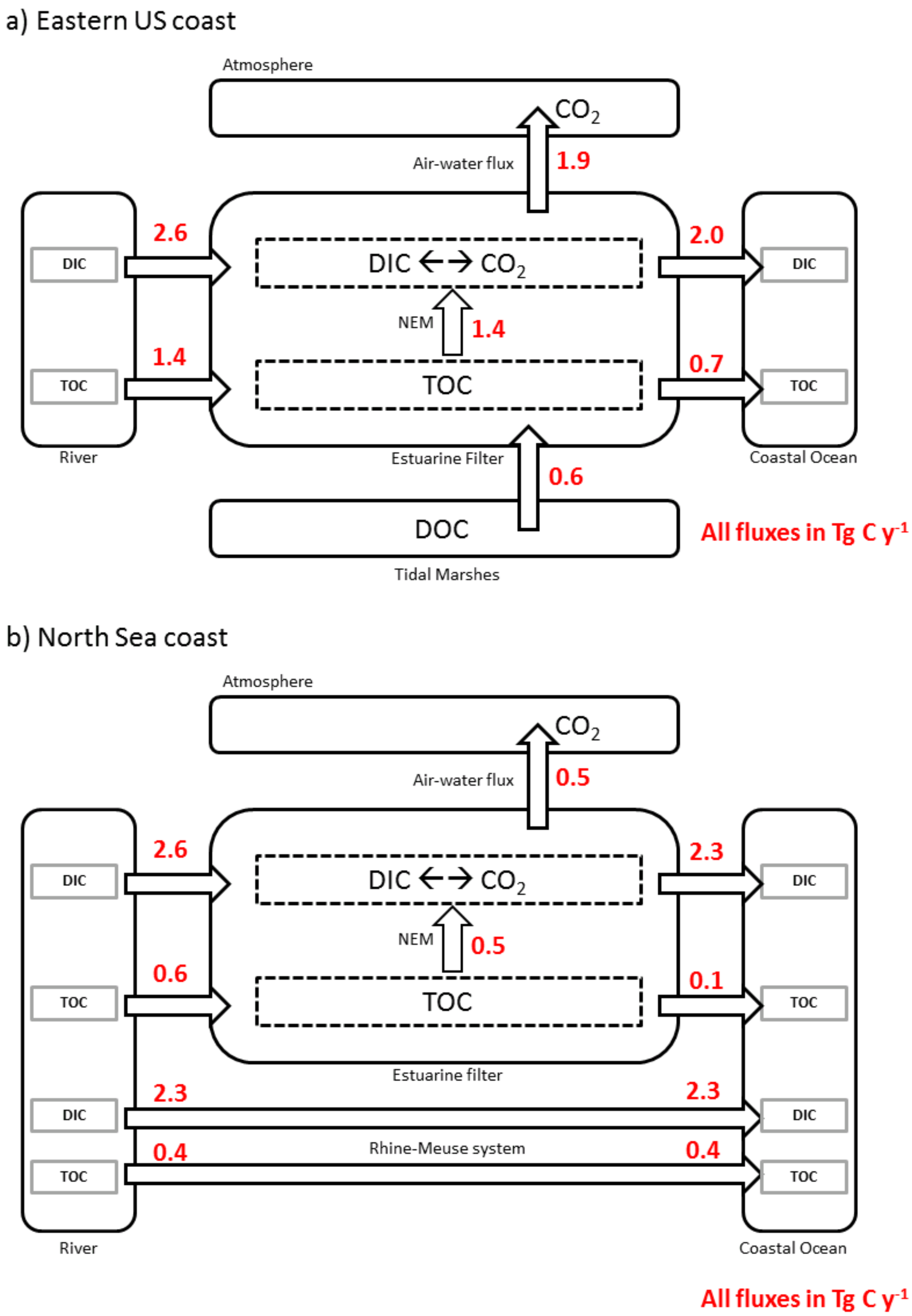

Figure 11: Annual carbon budget of the estuaries of the East coast of the US (a) and of the coast of the North Sea (b, modified from Volta et al., 2016a).

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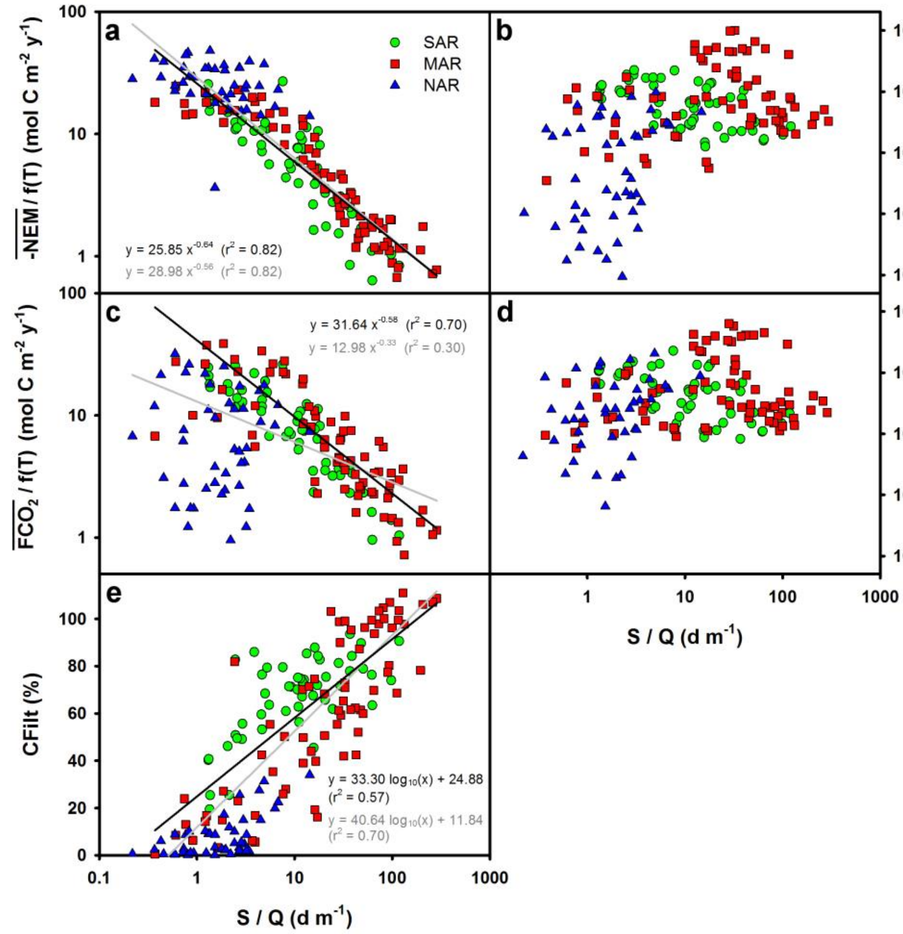
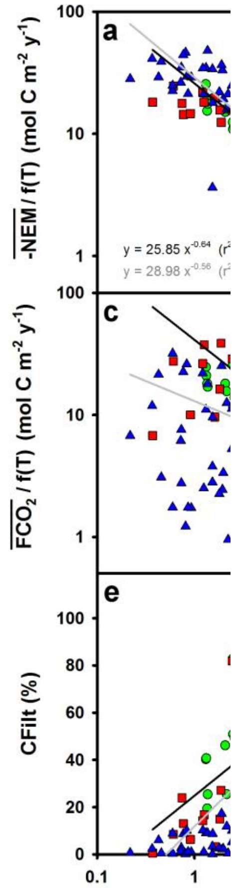


Figure 12: System scale integrated biogeochemical indicators expressed as functions of the depth normalized residence time expressed as the ratio of the estuarine surface S and the river discharge Q for all seasons. Panels b, d and e represent NEM, $-FCO_2$ and CFilt, respectively. Panels a and c represent NEM, $-FCO_2$ normalized by a temperature Q_{10} function. Black lines are the best fitted linear regressions obtained using all the point. Grey lines are best fit using only the estuaries from the MAR and SAR regions.



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Air-water CO_2 evasion from U.S. East Coast estuaries

Goossens, Nicolas[1], Laruelle, Goulven Gildas[1*], Arndt, Sandra[2], Cai, Wei-Jun[3] & Regnier, Pierre[1]

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Abstract:

This study presents the first regional-scale assessment of estuarine CO_2 evasion along the East coast of the US (25 – 45 °N). The focus is on 43 tidal estuaries, which together drain a catchment of 697000 km^2 or 76 % of the total area within this latitudinal band. The approach is based on the Carbon – Generic Estuarine Model (C-GEM) that allows simulating hydrodynamics, transport and biogeochemistry for a wide range of estuarine systems using readily available geometric parameters and global databases of seasonal climatic, hydraulic, and riverine biogeochemical information. Our simulations, performed using conditions representative of the year 2000, suggest that, together, US East coast estuaries emit 1.9 TgC yr^{-1} in the form of CO_2, which correspond to about 40 % of the carbon inputs from rivers, marshes and mangroves. Carbon removal within estuaries results from a combination of physical (outgassing of supersaturated riverine waters) and biogeochemical processes (net heterotrophy and nitrification). The CO_2 evasion and its underlying drivers show important variations across individual systems, but reveal a clear latitudinal pattern characterized by a decrease in the relative importance of physical over biogeochemical processes along a North-South gradient. Finally, the results reveal that the ratio of estuarine surface area to the river discharge, S/Q (which has a scale of per meter discharged water per year), could be used as a predictor of the estuarine carbon processing in future regional and global scale assessments.

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1 Introduction

Carbon fluxes along the land-ocean aquatic continuum are currently receiving increasing attention because of their recently recognized role in the global carbon cycle and anthropogenic CO_2 budget (Bauer et al., 2013; Regnier et al., 2013a; LeQuéré et al., 2014, 2015). Estuaries are important reactive conduits along this continuum, which links the terrestrial and marine global carbon cycles (Cai, 2011). Large amounts of terrestrial carbon transit through these systems, where they mix with carbon from autochthonous, as well as marine sources. During estuarine transit, heterotrophic processes degrade a fraction of the allochthonous and autochthonous organic carbon inputs, supporting a potentially significant, yet poorly quantified CO_2 evasion flux to the atmosphere. Recent estimates suggest that 0.15-0.25 PgC yr^{-1} is emitted from estuarine systems worldwide (Borges and Abril, 2012; Cai, 2011; Laruelle et al., 2010; Regnier et al., 2013a; Laruelle et al., 2013, Bauer et al., 2013). Thus, in absolute terms the global estuarine CO_2 evasion corresponds to about 15% of the open ocean CO_2 uptake despite the much smaller total surface area.

Currently, estimates of regional and global estuarine CO_2 emissions are mainly derived on the basis of data-driven approaches that rely on the extrapolation of a small number of local measurements (Cai, 2011; Chen et al., 2013; Laruelle et al., 2013). These approaches fail to capture the spatial and temporal heterogeneity of the estuarine environment (Bauer et al., 2013) and are biased towards anthropogenically influenced estuarine systems located in industrialized countries (Regnier et al., 2013a). Even in the best surveyed regions of the world (e.g. Australia, Western Europe, North America or China) observations are merely available for a small number of estuarine systems. In addition, if available, data sets are generally of low spatial and temporal resolution. As a consequence, data-driven approaches can only provide first-order estimates of regional and global estuarine CO_2 emissions.

Integrated model-data approaches can help here, as models provide the means to extrapolate over temporal and spatial scales and allow disentangling the complex and very dynamic network of

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physical and biogeochemical processes that controls estuarine CO_2 emissions. Over the past decades, increasingly complex process-based models have been applied, in combination with local data, to elucidate the coupled carbon-nutrient cycles on the scale of individual estuaries (e.g., O'Kane, 1980; Soetaert and Herman, 1995; Vanderborght et al., 2002; Lin et al., 2007; Arndt et al., 2009; Cerco et al., 2010; Baklouti et al., 2011). However, the application of such model approaches remains limited to the local scale due to their high data requirements for calibration and validation (e.g. bathymetric and geometric information and boundary conditions), as well as the high computational demand associated with resolving the complex interplay of physical and biogeochemical processes on the relevant temporal and spatial scales (Regnier et al., 2013b). Complex process-based models are thus not suitable for the application on a regional or global scale and, as a consequence, the estuarine carbon filter is, despite its increasingly recognized role in regional and global carbon cycling (e.g. Bauer et al., 2013), typically not taken into account in model-derived regional or global carbon budgets (Bauer et al., 2013). The lack of regional and global model approaches that could be used as stand-alone applications or that could be coupled to regional terrestrial river network models (e.g. GLOBALNEWS: Seitzinger et al., 2005; Mayorga et al., 2010; SPARROW: Schwarz et al., 2006) and continental shelf models (e.g. Hofmann et al., 2011) is thus critical.

The Carbon-Generic Estuary Model (C-GEM (v1.0); Volta et al., 2014) has been developed with the aim of providing such a regional/global modeling tool that can help improve existing, observationally derived first order estimates of estuarine CO_2 emissions. C-GEM (v1.0) has been specifically designed to reduce data requirements and computational demand and, thus, tackles the main impediments for the application of estuarine models on a regional or global scale. The approach takes advantage of the mutual dependency between estuarine geometry and hydrodynamics in alluvial estuaries and uses an idealized representation of the estuarine geometry to support the hydrodynamic calculations. It thus allows running steady state or fully transient annual to multi-decadal simulations for a large number of estuarine systems, using geometric information readily available through maps

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or remote sensing images. Although the development of such a regional/global tool inevitably requires simplification, careful model evaluations have shown that, despite the geometric simplification, C-GEM provides an accurate description of the hydrodynamics, transport and biogeochemistry in tidal estuaries (Volta et al., 2014). In addition, the model approach was successfully used to quantify the contribution of different biogeochemical processes for CO_2 air-water fluxes in an idealized, funnel-shaped estuary forced by typical summer conditions characterizing a temperate Western European climate (Regnier et al., 2013b). Volta et al. (2016b) further investigated the effect of estuarine geometry on the CO_2 outgassing using three idealized systems and subsequently established the first regional carbon budget for estuaries surrounding the North Sea by explicitly simulating the six largest systems of the area (Volta et al., 2016a), including the Scheldt and the Elbe for which detailed validation was performed.

Here, we extend the domain of application of C-GEM (v1.0) to quantify CO_2 exchange fluxes, as well as the overall organic and inorganic carbon budgets for the full suite of estuarine systems located along the entire East coast of the United States, one of the most intensively monitored regions in the world. A unique set of regional data, including partial pressure of CO_2 in riverine and continental shelf waters (pCO_2; Signorini et al., 2013; Laruelle et al., 2015), riverine biogeochemical characteristics (Lauerwald et al., 2013), estuarine eutrophication status (Bricker et al., 2007) and estuarine morphology (NOAA, 1985) are available. These comprehensive data sets are complemented by local observations of carbon cycling and CO_2 fluxes in selected, individual estuarine systems (see Laruelle et al., 2013 for a review), making the East coast of the United States an ideal region for a first, fully explicit regional evaluation of CO_2 evasion resolving every major tidal estuary along the selected coastal segment. The scale addressed in the present study is unprecedented so far (> 3000 km of coastline) and covers a wide range of estuarine morphological features, climatic conditions, land-use and land cover types, as well as urbanization levels. The presented study will not only allow a further evaluation of C-GEM (v1.0), but will also provide the first regional-scale assessment of estuarine CO_2 evasion along the East coast of the US (25 – 45 °N)

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and will help explore general relationships between carbon cycling and CO_2 evasion, and readily available estuarine geometrical parameters.

After a description of the model itself and of the dataset used to set up the simulations, a local validation is presented which includes salinity, pCO_2 and pH longitudinal profiles for two well monitored systems (the Delaware Bay and the Altamaha River Estuary). The yearly averaged rates of CO_2 exchange at the air-water interface simulated by the model for 13 individual estuaries are also compared with observed values reported in the literature. Next, regional scale simulations for 43 tidal estuaries of the eastern US coast provide seasonal and yearly integrated estimates of the Net Ecosystem Metabolism (NEM), CO_2 evasion and carbon filtering capacity, CFilt. Model results are then used to elucidate the estuarine biogeochemical behavior along the latitudinal transect encompassed by the present study (30-45° N). Finally, our results are used to derive general relationships between carbon cycling and CO_2 evasion, and readily available estuarine geometrical parameters.

2. Regional description and model approach

2.1 Observation-based carbon budget for the East coast of the United States

The study area covers the Atlantic coast of the United States (Fig.1), from the southern tip of Florida (25°N) to Cobscook Bay (45°N) at the US-Canada boundary. This area encompasses distinct climatic zones and land cover types and exhibits a variety of morphologic features (Fig. 1). The region can be subdivided into several sub-regions following a latitudinal gradient (Signorini et al., 2013). In this study, we define three sub-regions following the boundaries suggested by the COSCAT segmentation (Meybeck et al., 2006; Laruelle et al., 2013) and the further subdivision described in Laruelle et al. (2015). From North to South, the regions are called North Atlantic, Mid Atlantic and South Atlantic Regions (Fig. 1). Total carbon inputs from watersheds to US East coast estuaries (Tab. 1) have been

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estimated to range from 4.0 to 10.7 Tg C yr^{-1} (Mayorga et al., 2010; Shih et al., 2010; Stets and Strieg, 2012; Tian et al., 2010; Tian et al., 2012), consisting of dissolved organic carbon (DOC; ~50%), dissolved inorganic carbon (DIC; ~40%) and particulate organic carbon (POC; ~10%). In addition, a statistical approach has been applied to estuaries of the region to quantify organic carbon budgets and Net Ecosystem Productivity (NEP) using empirical models (Herrmann et al., 2015).

Recent studies estimated that, along the East coast of the United States, rivers emit 11.4 TgC yr^{-1} of CO_2 to the atmosphere (Raymond et al., 2013), while continental shelf waters absorb between 3.4 and 5.4 TgC yr^{-1} of CO_2 from the atmosphere (Signorini et al., 2013). A total of thirteen local, annual mean estuarine CO_2 flux estimates across the air-water interface based on measurements are also reported in the literature and are grouped along a latitudinal gradient (Tab. 2). Four of these estimates are located in the South Atlantic region (SAR): Sapelo Sound, Doboy Sound, Altamaha Sound (Jiang et al., 2008), and the Satilla River estuary (Cai and Wang, 1998). Three studies investigate CO_2 fluxes in the mid-Atlantic Region (MAR): the York River Estuary (Raymond et al., 2000) and the Hudson River (Raymond et al., 1997). There is also a comprehensive CO_2 flux study for the Delaware Estuary published after the completion of this work (Joeseof et al., 2015). Six systems are located in the North Atlantic region (NAR): The Great Bay, the Little Bay, the Oyster estuary, the Bellamy estuary, the Cocheco estuary (Hunt et al., 2010; 2011), and the Parker River estuary (Raymond and Hopkinson, 2003). The mean annual flux per unit area from these local studies is 11.7±13.1 mol C m^{-2} yr^{-1} and its extrapolation to the total estuarine surface leads to a regional CO_2 evasion estimate of 3.8 Tg C y^{-1}. This estimate is in line with that of Laruelle et al. (2013) for the same region which proposes an average CO_2 emission rate of 10.8 mol C m^{-2} yr^{-1}. Thus, CO_2 outgassing could remove 35% to 95% of the riverine carbon loads during estuarine transit. About 75 % of the air-water exchange occurs in tidal estuaries (2.8 Tg C y^{-1}) while lagoons and small deltas contribute to the remaining 25 %. Although these simple extrapolations from limited observational data are associated with large uncertainties, they highlight the potentially significant contribution of estuaries to the CO_2 outgassing in the region. However, process-based quantifications of regional organic and

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inorganic C budgets including air-water CO_2 fluxes for the estuarine systems along the East coast are not available.

2.2 Selection of estuaries

The National Estuarine Eutrophication Assessment (NEAA) survey (Bricker et al., 2007), which uses geospatial data from the National Oceanic and Atmospheric Administration (NOAA) Coastal Assessment Framework (CAF) (NOAA, 1985), was used to identify and characterize 58 estuarine systems discharging along the Atlantic coast of the United States. From this set, 43 'tidal' estuaries, defined as a river stretch of water that is tidally influenced (Dürr et al., 2011), were retained (Fig. 1) to be simulated by the C-GEM model, which is designed to represent such systems. Using outputs from terrestrial models (Hartmann et al., 2009; Mayorga et al., 2010), the cumulated riverine carbon loads for all the non-tidal estuaries that are excluded from the present study amount to 0.9 Tg C yr^{-1}, which represents less than 15% of the total riverine carbon loads of the region. These 15 systems are located in the SAR (10) and in the MAR (5).

The northeastern part of the domain (NAR, Fig. 1; Tab. 1) includes 20 estuaries along the Gulf of Maine and the Scotian shelf, covering a cumulative surface area of ~5300 km². It includes drowned valleys, rocky shores and a few tidal marshes. The climate is relatively cold (annual mean= 8°C) and the human influence is relatively limited because of low population density and low freshwater inputs. The mean estuarine water depth is 12.9 m and the mean tidal range is 2.8 m.

The central zone (MAR) includes 17 tidal estuaries accounting for a total surface area of 14500 km². The Chesapeake Bay and the Delaware estuaries alone contribute more than 60% to the surface area of the region. In this region, estuaries are drowned valleys with comparatively high river discharge and intense exchange with the ocean. Several coastal lagoons, characterized by a limited exchange with the ocean are located here, but are not included in our analysis. The Mid-Atlantic Region (MAR) is characterized by a mean annual temperature of 13°C and is strongly impacted by human activities,

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due to the presence of several large cities (e.g. New York, Washington, Philadelphia, Baltimore) and intense agriculture. The mean water depth is about 4.7 m and the tidal range is 0.8 m.

The southern Atlantic region (SAR) includes 10 tidal estuaries covering a total surface area of 12182 km^2. These systems are generally dendritic and surrounded by extensive salt marshes. The climate is subtropical with an average annual temperature of 19°C. Land use includes agriculture and industry, but the population density is generally low. Estuarine systems in the SAR are characterized by a shallow mean water depth of 2.9 m and a tidal range of 1.2 m.

2.3 Model set-up

The generic 1D Reactive-Transport Model (RTM) C-GEM (Volta et al., 2014) is used to quantify the estuarine carbon cycling in the 43 systems considered in this study. The approach is based on idealized geometries (Savenije, 2005; Volta et al., 2014) and is designed for regional and global scale applications (Regnier et al., 2013b; Volta et al., 2014, 2016a). The model approach builds on the premise that hydrodynamics exerts a first-order control on estuarine biogeochemistry (Arndt et al., 2007; Friedrichs and Hofmann, 2001) and CO_2 fluxes (Regnier et al., 2013a). The method takes advantage of the mutual dependence between geometry and hydrodynamics in tidal estuaries (Savenije, 1992) and the fact that, as a consequence, transport and mixing can be easily quantified from readily available geometric data (Regnier et al., 2013a; Savenije, 2005; Volta et al., 2016b).

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2.3.1 Description of idealized geometries for tidally-averaged conditions

Although tidal estuaries display a wide variety of shapes, they nevertheless share common geometric characteristics that are compatible with an idealized representation (Fig. 2, Savenije, 1986; Savenije, 2005). For tidally-averaged conditions, their width B (or cross-sectional area A) can be described by an exponential decrease as a function of distance, *x*, from the mouth (Savenije, 1986; Savenije, 2005):

$$B = B0 * \exp\left(-\frac{x}{b}\right) \qquad (1)$$

where B (m) is the tidally averaged width, B0 (m) the width at the mouth, x (m) the distance from the mouth (x=0) and b (m) the width convergence length (Fig. 2). The width convergence length, b, is defined as the distance between the mouth and the point at which the width is reduced to B0 e^{-1}. It is directly related to the dominant hydrodynamic forcing. A high river discharge typically results in a prismatic channel with long convergence length (river dominated estuary), while a large tidal range results in a funnel-shaped estuary with short convergence length (marine dominated estuary). At the upstream boundary, the estuarine width is given by:

$$B_L = B0 * \exp\left(-\frac{L}{b}\right) \qquad (2)$$

Where L denotes the total estuarine length (m) along the estuarine longitudinal axis.

The total estuarine surface S (m^2) can be estimated by integrating equation (1) over the estuarine length:

$$S = \int_0^L B\,dx = b * B0 * \left(1 - \exp\left(-\frac{L}{b}\right)\right) \qquad (3)$$

The width convergence length is then calculated from B0, B_L, L and the real estuarine surface area (SR) by inserting equation (2) in equation (3):

$$b = \frac{SR}{B0 - BL} \qquad (4)$$

SR is calculated for each system using the SRTM water body data (Fig. 3a), a geographical dataset encoding high-resolution worldwide coastal outlines in a vector format (NASA/NGA, 2003). While such a database exists for a well monitored region such as the East coast of the US, resorting to using the idealized estuarine surface area (S) is necessary in many other regions. The longitudinal

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mean, tidally averaged, depth h (m), is obtained from the National Estuarine Eutrophication Assessment database (Bricker et al., 2007).

Using this idealized representation, the estuarine geometry can be defined by a limited number of parameters: the width at the mouth (B_0), the estuarine length (L), the estuarine width at the upstream limit (B_L) and the mean depth h. These parameters can be easily determined from local maps or Google Earth using Geographic Information Systems (GIS) or obtained from databases (NASA/NGA, 2003).

2.3.2 Hydrodynamics, transport and biogeochemistry

Estuarine hydrodynamics are described by the one-dimensional barotropic, cross-sectionally integrated mass and momentum conservation equations for a channel with arbitrary geometry (Nihoul and Ronday, 1976; Regnier et al., 1998; Regnier and Steefel, 1999):

$$r_s \frac{\partial A}{\partial t} + \frac{\partial Q}{\partial x} = 0 \qquad (5)$$

$$\frac{\partial U}{\partial t} + U\frac{\partial U}{\partial x} = -g\frac{\partial \zeta}{\partial x} - g\frac{U|U|}{C_z^2 H} \qquad (6)$$

where:

t	time	[s]
x	distance along the longitudinal axis	[m]
A	cross-section area $A = H \cdot B$	[m^2]
Q	cross-sectional discharge $Q = A \cdot U$	[$m^3\ s^{-1}$]
U	flow velocity Q / A	[$m\ s^{-1}$]
r_s	storage ratio $r_s = B_s / B$	[-]

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B_s	storage width	[m]
g	gravitational acceleration	[m s^{-2}]
ξ	elevation	[m]
H	total water depth $H = h + \xi(x,t)$	[m]
C_z	Chézy coefficient	[m$^{1/2}$ s^{-1}]

The coupled partial differential equations (Eqs. (5) and (6)) are solved by specifying the elevation ξ_0(t) at the estuarine mouth and the river discharge Q_r(t) at the upstream limit of the model domain.

The one-dimensional, tidally-resolved, advection-dispersion equation for a constituent of concentration *C(x,t)* in an estuary can be written as (e.g. Pritchard, 1958):

$$\frac{\partial C}{\partial t} + \frac{Q}{A}\frac{\partial C}{\partial x} = \frac{1}{A}\frac{\partial}{\partial x}\left(AD\frac{\partial C}{\partial x}\right) + P \qquad (7)$$

where *Q*(x,t) and *A*(x,t) denote the cross-sectional discharge and area, respectively and are provided by the hydrodynamic model (eq. 5 and 6). *P*(x,t) is the sum of all production and consumption process rates affection the concentration of the constituent. The effective dispersion coefficient *D* (m^2 s^{-1}) implicitly accounts for dispersion mechanisms associated to sub-grid scale processes (Fischer, 1976; Regnier et al., 1998). In general, *D* is maximal near the sea, decreases upstream and becomes virtually zero near the tail of the salt intrusion curve (Preddy, 1954; Kent, 1958; Ippen and Harleman, 1961; Stigter and Siemons, 1967). The effective dispersion at the estuarine mouth can be quantified by the following relation (Savenije, 1986):

$$D_0 = 26 \cdot (h_0)^{1.5} \cdot (N \cdot g)^{0.5} \qquad (8)$$

where h_0 (m) is the tidally-averaged water depth at the estuarine mouth and *N* is the dimensionless Canter Cremers' estuary number defined as the ratio of the freshwater entering the estuary during a

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tidal cycle to the volume of salt water entering the estuary over a tidal cycle (Simmons, 1955).

$$N = \frac{Q_b \cdot T}{P} \qquad (9)$$

In this equation, Q_b is the bankfull discharge ($m^3\ s^{-1}$), T is the tidal period (s) and P is the tidal prism (m^3). For each estuary, N can thus be calculated directly from the hydrodynamic model. The variation in *D* along the estuarine gradient can be described by Van der Burgh's equation (Savenije, 1986):

$$\frac{\partial D}{\partial x} = -K\frac{Q_r}{A} \qquad (10)$$

where *K* is the dimensionless Van der Burgh's coefficient and the minus sign indicates that *D* increases in downstream direction (Savenije, 2012). The Van der Burgh's coefficient is a shape factor that has values between 0 and 1 (Savenije, 2012), and is a function of estuarine geometry for tidally average conditions. Therefore, each estuarine system has its own characteristic *K* value, which correlates with geometric and hydraulic scales (Savenije, 2005). Based on a regression analysis covering a set of 15 estuaries, it has been proposed to constrain *K* from the estuarine geometry (Savenije, 1992):

$$K = 4.32 \cdot \frac{h_0^{0.36}}{B_0^{0.21} \cdot b^{0.14}} \quad \text{with} \quad 0 < K < 1 \qquad (11)$$

Reaction processes *P* considered in C-GEM comprise aerobic degradation, denitrification, nitrification, primary production, phytoplankton mortality and air-water gas exchange for O_2 and CO_2 (Fig. 4 and Tab. 3). These processes and their mathematical formulation are described in detail in Volta et al. (2014) and Volta et al. (2016a).

The non-linear partial differential equations for the hydrodynamics are solved by a finite difference scheme following the approach of (Regnier et al., 1997; Regnier and Steefel, 1999) and (Vanderborght et al., 2002). The timestep Δt is 150s and the grid size Δx is constant along the

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longitudinal axis of the estuary. The grid size default value is 2000 m, but can be smaller for short length estuaries to guarantee a minimum of 20 grid points within the computational domain. Transport and reaction terms are solved in sequence within a single timestep using an operator splitting approach (Regnier et al., 1997). The advection term in the transport equation is integrated using a third-order accurate total variation diminishing (TVD) algorithm with flux limiters (Regnier et al., 1998), ensuring monotonicity (Leonard, 1984), while a semi-implicit Crank-Nicholson algorithm is used for the dispersion term (Press et al., 1992). These schemes have been extensively tested using the CONTRASTE estuarine model (e.g. Regnier et al., 1998; Regnier and Steefel, 1999; Vanderborght et al., 2002) and guarantee mass conservation to within <1%. The reaction network (including erosion-deposition terms when the constituent is a solid species), is numerically integrated using the Euler method (Press et al., 1992). The primary production dynamics, which requires vertical resolution of the photic depth, is calculated according to the method described in Vanderborght et al. (2007). This method assumes an exponential decrease of the light in the water column (Platt et al., 1980), which is solved using a Gamma function.

2.4 Boundary and forcing conditions

Boundary and forcing conditions are extracted from global databases and global model outputs that are available at 0.5° resolution. Therefore, C-GEM simulations are performed at the same resolution according to the following procedure. First, 43 coastal cells corresponding to tidal estuaries are identified in the studied area (Fig. 1). If the mouth of an estuary is spread over several 0.5° grid cells, those cells are regrouped in order to represent a single estuary (e.g. Delaware estuary), and subsequently, a single idealized geometry is defined as described above. The model outputs (Hartmann et al. , 2009; Mayorga et al., 2010) and databases (Antonov et al., 2010; Garcia et al., 2010a; Garcia et al., 2010b) used to constrain our boundary conditions are representative of the year 2000.

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For each resulting cell, boundary and forcing conditions are calculated for the following periods: January-March; April-June; July-September and October-December. This allows for an explicit representation of the seasonal variability in the simulations.

2.4.1 External forcings

Transient physical forcings are calculated for each season and grid cell using monthly mean values of water temperature (World Ocean Atlas, 2009) and seasonal averaged values for wind speed (Cross-Calibrated-Multi-Platform (CCMP) Ocean Surface Wind Vector Analyses project (Atlas et al., 2011)). Mean daily solar radiation and photoperiods (corrected for cloud coverage using the ISCCP Cloud Data Products, Rossow and Schiffer, 1999) are calculated depending on latitude and day of the year using a simple model (Brock, 1981).

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2.4.2 Riverine discharge, concentrations and fluxes

River discharges are extracted from the UNH/GRDC runoff dataset (Fekete et al., 2002). These discharges represent long-term averages (1960-1990) of monthly and annual runoff at 0.5 degree resolution. The dataset is a composite of long-term gauging data, which provides average runoff for the largest river basins, and a climate driven water balance model (Fekete et al., 2002). Total runoff values are then aggregated for each watershed at the coarser 0.5 degree resolution (Fig. 3b). Next, seasonal mean values (in m^3 s^{-1}) are derived in order to account for the intra-annual variability in water fluxes. Based on annual carbon and nutrients inputs from the watersheds (Mg y^{-1}), mean annual concentrations (mmol m^{-3}) are estimated for each watershed using the UNH/GRDC annual runoff (km^3 y^{-1}). Mean seasonal concentrations are then calculated from the seasonally resolved river water fluxes of a given sub-region.

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Annual inputs of dissolved organic carbon (DOC), particulate organic carbon (POC) and inorganic nutrients are derived from the globalNEWS2 model (Mayorga et al., 2010). Global NEWS is a spatially explicit, multi-element (N, P, Si, C) and multi-form global model of nutrient exports by rivers. In a nutshell, DOC exports are a function of runoff, wetland area, and consumptive water use (Harrison et al., 2005). No distinction is made between agricultural and natural landscapes, since they appear to have similar DOC export coefficients (Harrison et al., 2005). Sewage inputs of OC are ignored in GlobalNEWS, because their inclusion did not improve model fit to data (Harrison et al., 2005). POC exports from watersheds are estimated using an empirical relationship with Suspended Particulate Matter (SPM; Ludwig et al., 1996). Inorganic nitrogen (DIN) and phosphorus (DIP) fluxes calculated by GlobalNEWS depend on agriculture and tropical forest coverage, fertilizer application, animal grazing, sewage input, atmospheric N deposition and biological N fixation (Mayorga et al., 2010). The inputs of dissolved silica (DSi) are controlled by soil bulk density, precipitation, slope, and presence of volcanic lithology (Beusen et al., 2009).

The DIN speciation is not provided by the GlobalNEWS2 model. The NH_4 and NO_3 concentrations are therefore determined independently on the basis of an empirical relationship between ammonium fraction (NH4/DIN ratio) and DIN loads (Meybeck, 1982). Dissolved Oxygen (DO) concentrations are extracted from the water quality criteria recommendations published by the United States Environmental Protection Agency (EPA, 2009). The same source is used for phytoplankton concentrations, using a chlorophyll-a to phytoplankton carbon ratio of 50 gC $(gChla)^{-1}$ (Riemann et al., 1989) to convert the EPA values to carbon units used in the present study.

Inputs of dissolved inorganic carbon (DIC) and total Alkalinity (ALK) are calculated from values reported in the GLORICH database (Hartmann et al., 2009). For each watershed, seasonal mean values of DIC and ALK concentrations are estimated from measurements performed at the sampling locations that are closest to the river-estuary boundary. The spatial distribution of annual inputs of TOC=DOC+POC, DIC, and TC=TOC+DIC from continental watersheds to estuaries are reported in Fig. 5a, 5c and 5d, respectively. The contribution of tidal wetlands to the TOC inputs is also shown (Fig. 5b). Overall, the TC input over the entire model domain is estimated at 4.6 Tg C yr^{-1}, which falls in the lower end of previous reported estimations (Najjar et al. 2012).

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2.4.3 Inputs from tidal wetlands

The DOC input of estuarine wetlands (Fig. 5b) scales to their fraction, W, of the total estuarine and is calculated using the GlobalNEWS parameterization:

$$Y\_DOC = \frac{\left[(E\_C_{wet} * W) + E\_C_{dry} * (1 - W)\right] * R^{a} * Q_{act}}{Q_{nat}} \qquad (12)$$

$$\frac{Y\_DOC_{wet}}{Y\_DOC} = \frac{E\_C_{wet} * W}{E\_C_{wet} * W + E\_C_{dry} * (1 - W)} \qquad (13)$$

where Y_DOC is the DOC yield (kg C km^{-2} y^{-1}) calculated for the entire watershed, Y_DOC_{wet} is the estimated DOC yield from wetland areas (kg C km^{-2} y^{-1}), Q_{act}/Q_{nat} is the ratio between the measured discharge after dam construction and before dam construction, E_C_{wet} and E_C_{dry} (kg C km^{-2} y^{-1}) are the export coefficients of DOC from wetland and non-wetland soils, respectively. W is the percentage of the land area within a watershed that is covered by wetlands, R is the runoff (m y^{-1}) and *a* is a unit-less calibration coefficient defining how non-point source DOC export responds to runoff. The value of a is set to 0.95, consistent with the original GlobalNEWS -DOC model of Harrison et al. (2005). The carbon load Y_DOC_{wet} is then exported as a diffuse source along the relevant portions of estuary. The estuarine segments receiving carbon inputs from tidal wetlands are identified using the National Wetlands Inventory of the U.S. Fish and Wildlife Service (U.S. Fish and Wildlife Service, 2014). The inputs from those systems are then allocated to the appropriate grid cell of the model domain using GIS. The flux calculated is an annual average that is subsequently partitioned between the four seasons as a function of the mean seasonal temperature, assumed to be the main control of the wetland-estuarine exchange. This procedure reflects the observation that

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in spring and early summer, DOC export is small as a result of its accumulation in the salt marshes induced by the high productivity (Dai and Wiegert, 1996), (Jiang et al., 2008). In late summer and fall, the higher water temperature and greater availability of labile DOC contribute to higher bacterial remineralization rates in the intertidal marshes (Cai et al., 1999; Middelburg et al., 1996; Wang and Cai, 2004), which induce an important export. This marsh production-recycle-export pattern is consistent with the observed excess DIC signal in the offshore water (Jiang et al. 2013). DIC export from tidal wetlands is neglected here because it is assumed that OC is not degraded before reaching the estuarine realm. Although this assumption may lead to an overestimation of OC export from marshes and respiration in estuarine water, it will not significantly affect the water pCO_2 and degassing in the estuarine waters because mixing is faster than respiration.

2.4.4 Concentrations at the estuarine mouth

For each estuary, the downstream boundary is located 20 km beyond the mouth to minimize the bias introduced by the choice of a fixed concentration boundary condition to characterize the ocean water masses (e.g. Regnier et al., 1998). This approach also reduces the influence of marine boundary conditions on the simulated estuarine dynamics, especially for all organic carbon species whose concentrations are fixed at zero at the marine boundary. This assumption ignores the intrusion of marine organic carbon into the estuary during the tidal cycle but allows focusing on the fate of terrigenous material and its transit through the estuarine filter. DIC concentrations are extracted from the GLODAP dataset (Key et al., 2004), from which ALK and pH are calculated assuming CO_2 equilibrium between coastal waters and the atmosphere. The equilibrium value is computed using temperature (WOA2009, Locarnini et al., 2010) and salinity (WOA2009, Antonov et al. (2010)) data which vary both spatially and temporally. The equilibrium approach is a reasonable assumption because differences in partial pressure ΔpCO_2 between coastal waters and the atmosphere are generally much smaller (0-250 µatm (Signorini et al., 2013)) than those reported for estuaries (ΔpCO_2 in the range 0-10000 µatm (Borges and Abril, 2012)). Salinity, DO, NO_3, DIP and DSi

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concentrations are derived from the World Ocean Atlas (Antonov et al., 2010; Garcia et al., 2010a; Garcia et al., 2010b). NH_4 concentrations are set to zero in marine waters. For all variables, seasonal means are calculated for each grid cell of the boundary.

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2.5 Biogeochemical indicators

The model outputs (longitudinal profiles of concentration and reaction rates) are integrated in time over the entire volume or surface of each estuary to produce the following indicators of the estuarine biogeochemical functioning (Regnier et al., 2013b): the mean annual Net Ecosystem Metabolism (*NEM*), the air-water CO_2 flux (FCO_2), the carbon and nitrogen filtering capacity (*CFilt* and *NFilt*) and their corresponding element budgets. The *NEM* (molC y^{-1}) (Caffrey, 2004; Odum, 1956) is defined as the difference between net primary production (*NPP*) and total heterotrophic respiration (*HR*) at the system scale:

$$NEM = \int_0^{365} \int_0^{L} [NPP(x,t) - R_{aer}(x,t) - R_{den}(x,t)] * B(x) * H(x,t)\, dx\, dt \qquad (14)$$

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where *NPP* is the Net Primary Production (mol C m^{-3} y^{-1}), R_{aer} the aerobic degradation of organic matter (in mol C m^{-3} y^{-1}) and R_{den} the denitrification (in mol C m^{-3} y^{-1}) (see Volta et al., 2014 for detailed formulations). *NEM* is thus controlled by the production and decomposition of autochthonous organic matter, by the amount and degradability of organic carbon delivered by rivers and tidal wetlands and by the export of terrestrial and in-situ produced organic matter to the adjacent coastal zone. Following the definition of *NEM*, the trophic status of estuaries can be net heterotrophic (NEM<0) when *HR* exceeds *NPP* or net autotrophic (NEM>0), when *NPP* is larger than *HR* because the burial and export of autochthonous organic matter exceeds the decomposition of river-borne material.

The FCO_2 (mol C y^{-1}) is defined as:

$$FCO_2 = \int_0^{365} \int_0^{L} RCO_2(x,t) * B(x)\, dx\, dt \qquad (15)$$

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$$RCO_2(x,t) = -\mathrm{v_p}(\mathrm{x,t})\left(\left[CO_{2(aq)}\right](x,t) - K_0(x,t) * P_{CO2}(x,t)\right) \qquad (16)$$

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where RCO_2 (molC m^{-2} y^{-1}) is the rate of exchange in CO_2 at the air-water interface per unit surface area, v_p is the piston velocity (m y^{-1}) and is calculated according to Regnier et al. (2002) to account for the effect of current velocity and wind speed, [*CO2(aq)*] is the concentration of CO_2 in the estuary (mol m^{-3}), K_0 is Henry's constant of CO_2 in sea water (mol m^{-3} atm^{-1}) and P_{CO2} is the atmospheric partial pressure in CO_2 (atm).

The carbon filtering capacity (in %) corresponds to the fraction of the river-borne supply that is lost to the atmosphere and is defined here as the ratio of the net outgassing flux of CO_2 and the total inputs of C, e.g. total carbon expressed as the sum of inorganic and organic carbon species, both in the dissolved and particulate phases.

$$CFilt = \frac{FCO_2}{\int_0^{365} Q * [TC]_{riv}\, dt} * 100 \qquad (17)$$

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where $[TC]_{riv}$ denote the total concentrations of C in the riverine inputs.

Fluxes per unit area for FCO_2 and *NEM*, noted $\overline{FCO_2}$ and $\overline{NEM}$, respectively, are defined in mol C m^{-2} y^{-1} and are calculated by dividing the integrated values calculated above by the (idealized) estuarine surface *S*:

$$\overline{NEM} = \frac{NEM}{S} * 1000 \qquad (18)$$

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$$\overline{FCO_2} = \frac{FCO_2}{S} * 1000 \qquad (19)$$

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Seasonal values for the biogeochemical indicators are calculated using the same formula as above, but calculate the integral over a seasonal rather than annual timescale (i.e. 3 months).

2.6 Model-data comparison

C-GEM has been specifically designed for an application on a global/regional scale requiring the representation of a large number of individual and often data-poor systems. Maximum model transferability and minimum validation requirements were thus central to the model design process and the ability of the underlying approach in reproducing observed dynamics with minimal calibration effort has been extensively tested. The performance C-GEM's one-dimensional hydrodynamic and transport models using idealized geometries have been evaluated for a number of estuarine systems exhibiting a wide variety of shapes (Savenije, 2012). In particular, it has been shown that the estuarine salt intrusion can be successfully reproduced using the proposed modeling approach (Savenije 2005; Volta et al., 2014; 2016b). In addition, C-GEM's biogeochemistry has also been carefully validated for geometrically contrasting estuarine system in temperate climate zones. Simulations for the Scheldt Estuary (Belgium and the Netherlands), a typical funnel-shaped estuary, were validated through model-data and model-model comparison (Volta et al., 2014; Volta et al., 2016a). Furthermore, simulations for the Elbe estuary (Germany), a typical prismatic shape estuary that drains carbonate terrains and, thus, exhibits very high pH was validated against field data (Volta et al., 2016a). In addition, C-GEM carbon budgets have been compared budget derived from observations for 6 European estuaries discharging in the North Sea (Volta et al., 2016a). Although C-GEM has been specifically designed and tested for the type of regional application presented here, its transferability from North Sea to US East Coast estuaries was further evaluated by assessing its performance in two East Coast estuaries. First, the hydrodynamic and transport model was tested

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for the Delaware Bay (MAR). The model was forced with the monthly, minimal and maximal observed discharge at Trenton over the period between 1912 and 1985 (UNH/GRDC Database, Fekete et al., 2000). Simulated salinity profiles are compared with salinity observations from January, February, May and June (the months with the highest number of data entries), which were extracted from the UNH/GRDC Database. Figure 6 shows that the model captures both the salinity intrusion length and the overall shape of the salinity profile well. In addition, the performance of the biogeochemical model and specifically its ability to reproduce pH and pCO_2 profiles was evaluated by a model-data comparison for both the Delaware Bay (MAR) in July 2003 and the Altamaha river estuary (SAR) in October 1995. Similar to Volta et al., 2016a, the test systems were chosen due to their contrasting geometries. The Delaware Bay is a marine dominated system characterized by a pronounced funnel shape, while the Altamaha River has a prismatic estuary characteristic of river dominated systems (Jiang et al., 2008). Monthly upstream boundary conditions for nutrients, as well as observed pH data and calculated pCO_2 are extracted from datasets described in (Sharp, 2010) and (Sharp et al., 2009) for the Delaware and in (Cai and Wang, 1998; Jiang et al., 2008) and (Cai et al., 1998) for the Altamaha river estuary. The additional forcings and boundary conditions are set similarly to the simulation for 2000 (see Tab. 2, 3, 4, 5, 6 in SI). Figure 7 shows that measured and simulated pH values are in good agreement with observed pH and observation-derived calculations of pCO_2. In the Delaware Bay, a pH minimum is located around km 140 and is mainly caused by intense nitrification sustained by large inputs of NH_4 from the Philadelphia urban area, coupled to an intense heterotrophic activity. Both processes lead to a well-developed pCO_2 increase in this area (Fig. 7b). Although no pCO_2 data were available for validation for the period from which boundary conditions were extracted, the simulated profile agree with pCO_2 measurement from July 2013 presented by Joesoef et al. (2015) with pCO_2 values close to equilibrium with the atmosphere in the widest section of the Delaware Bay (close to the estuarine mouth) and values above 1200 µatm at salinities below 5. For the Altamaha river estuary, pH steadily increases from typical river to typical coastal ocean values (Fig. 7b). In addition, both observations and model results reveal that

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outgassing is very intense in the low-salinity region with more than a 5 fold decrease in pCO_2 between salinity 0 and 5 (Fig. 7d).

While such local validations allow assessing the performance of the model for a specific set of conditions, the purpose of this study is to capture the average biogeochemical behavior of the estuaries of the eastern coast of the US. Therefore, in addition to the system-specific validation, published annually averaged FCO_2 estimates for 13 tidal systems located within the study area collected over the 1994-2006 period are compared to simulated FCO_2 for conditions representative of the year 2000. Overall, simulated FCO_2 are comparable to values reported in the literature (Tab. 2). Although discrepancies, which sometimes can significant, are observed at the level of individual systems, the model captures remarkably well the overall trend in CO_2 evasion rate across estuaries. The model simulates low CO_2 efflux (< 5 mol C m^{-2} yr^{-1}) for the 7 systems were such conditions have been observed, while the 6 systems for which the CO_2 evasion exceeds 10 mol C m^{-2} yr^{-1} are the same in the observations and in the model runs. The discrepancy at the individual system level likely result from a combination of factors, including the choice of model processes and there parametrization, the uncertainties in constraining boundary conditions and the limited representability of instantaneous and local observed.

3 Results and discussion

3.1 Spatial variability of estuarine carbon dynamics

Figure 8 presents the spatial distribution of simulated mean annual $\overline{FCO_2}$ and -$\overline{NEM}$ (Fig. 8a), as well as FCO_2 and -*NEM* (Fig. 8b). In general, mean annual $\overline{FCO_2}$ are about 30% larger than mean annual $\overline{NEM}$, with the exception of six estuaries situated in the North of the coastal segment. Overall, the $\overline{NEM}$ is characterized by smaller system to system variability compared to the $\overline{FCO_2}$ in all regions. In addition, Fig. 8 reveals distinct differences across the three coastal segments and highlights the

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important influence of the estuarine geometry and residence time, as well as the latitudinal temperature gradient on estuarine carbon cycling.

Overall, $\overline{FCO_2}$ values are the lowest in the NAR (mean flux = 17.3 ± 16.4 mol C m^{-2} y^{-1}; surface weighted average = 23.1 mol C m^{-2} y^{-1}), consistent with previously reported very low values for small estuaries surrounding the Gulf of Maine (Hunt et al., 2010; 2011; Tab. 2). In contrast, $\overline{NEM}$ reveals a regional minimum in the NAR (-51.2 ± 16.6 mol C m^{-2} y^{-1}; surface weighted average = -52.8 mol C m^{-2} y^{-1}). The MAR is characterized by intermediate values for $\overline{FCO_2}$, with a mean flux of 26.3 ± 34.6 mol C m^{-2} y^{-1} (surface weighted average =11.1 mol C m^{-2} y^{-1}) and lowest values for $\overline{NEM}$ (-15.1 ± 14.2 mol C m^{-2} y^{-1}; surface weighted average =-7.4 mol C m^{-2} y^{-1}). This region also shows the largest variability in CO_2 outgassing compared to the NAR and SAR, with the standard deviation exceeding the mean $\overline{FCO_2}$, and individual estimates ranging from 3.9 mol C m^{-2} y^{-1} to 150.8 mol C m^{-2} y^{-1}. This variability is mainly the result of largely variable estuarine surface areas and volumes. Some of the largest East coast estuaries (e.g. Chesapeake and Delaware Bays), as well as some of smallest estuaries (e.g. York River and Hudson River estuaries, Raymond et al., 1997; 2000), are located in this region (Tab. 2 and 4). The maximum values of 150.8 mol C m^{-2} y^{-1} simulated in the MAR are similar to the highest FCO_2 reported in the literature (132.3 mol C m^{-2} y^{-1} for the Tapti estuary in India; Sarma et al., 2012). The SAR is characterized by the highest mean $\overline{FCO_2}$ (46.7 ± 33.0 mol C m^{-2} y^{-1}; surface weighted average = 40.0 mol C m^{-2} y^{-1}) and intermediate $\overline{NEM}$ (-36.8 ± 24.7 mol C m^{-2} y^{-1}; surface weighted average = -31.2 mol C m^{-2} y^{-1}).

The NAR is characterized by a regional minimum in $\overline{FCO_2}$, and only contributes 4.6% to the total FCO_2 of the East coast of the US, owing to the small cumulative surface area available for gas exchange in its 10 estuarine systems. In contrast, the 18 MAR estuaries, with their large relative contribution to the total regional estuarine surface area, account for as much as 70.1% of the total outgassing. Because of their smaller cumulated surface area compared to those of the MAR, the 14 SAR estuaries account for merely 25.3% of the total outgassing despite their regional maximal $\overline{FCO_2}$.

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A similar, yet slightly less pronounced pattern emerges for the $\overline{NEM}$. The NAR, MAR and SAR respectively contribute 13.7%, 60.7% and 25.6% to the total regional net ecosystem metabolism. The comparatively larger relative contribution of the NAR to the total *NEM* as compared to the total FCO_2 can be explained by the importance of the specific aspect ratio for NEM. A larger ratio of estuarine width b0 and convergence length b corresponds to a more funnel shaped estuary while a low ratio corresponds to a more prismatic geometry (Savenije, 2000; Volta et al., 2014). In the NAR, estuaries are generally characterized by relatively narrow widths and deep-water depths, thus limiting the potential surface area for gas exchange with the atmosphere. However, the relative contribution of each region to the total regional *NEM* and FCO_2 is largely controlled by estuarine surface area. Figure 9 illustrates the cumulative *NEM* (a) and FCO_2 (b) as a function of the cumulative estuarine surface areas. The disproportionate contribution of large estuaries from the MAR translates into a handful of systems (Chesapeake and Delaware Bays and the main tributaries of the former, in particular) contributing to roughly half of the regional *NEM* and FCO_2, in spite of relatively low individual rates per unit surface area. However, the smallest systems (mostly located in the NAR and SAR) nevertheless still contribute a significant fraction to the total regional *NEM* and FCO_2. The 27 smallest systems merely account for less than 10% of the total regional estuarine surface area, yet contribute 38% and 29% to the total regional *NEM* and FCO_2, respectively (Fig. 9). This disproportioned contribution can be mainly attributed to their high individual $\overline{FCO_2}$ and $\overline{NEM}$. This is illustrated by the average simulated $\overline{FCO_2}$ for all 27 smallest systems (calculated as the sum of each estuarine CO_2 outgassing per unit surface area divided by the total number of estuarine systems) which is significantly higher (30.2 mol C m^{-2} y^{-1}) than its surface weighted average (14 mol C m^{-2} y^{-1}). Thereby accounting for the disproportionate contribution of very large systems (calculated as the sum of each estuarine CO_2 outgassing divided by the total estuarine surface area across the region).

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Following the approach used in Regnier et al. (2013), the contribution of each biogeochemical process to FCO_2 is assessed by evaluating their individual contribution to DIC and ALK changes taking into account the local buffering capacity of an ionic solution when TA and DIC are changing due to internal processes, but ignoring advection and mixing (Zeebe and Wolf-Gladrow 2001). In the present study, we quantify the effect of the NEM on the CO_2 balance, which is almost exclusively controlled by aerobic degradation rates because the contributions of denitrification and NPP to the net ecosystem balance are small. Nitrification, a process triggered by the transport and/or production of NH_4 in oxygenated waters, favors outgassing through its effect on pH, which shifts the acid-base equilibrium of carbonate species and increases the CO_2 concentration. The contribution of supersaturated riverine waters to the overall estuarine CO_2 dynamics is calculated as difference between all the other processes creating or consuming CO_2. Figure 10a presents the contribution of the annually integrated *NEM*, nitrification and evasion of supersaturated, DIC enriched riverine waters to the total outgassing for each system, as well as for individual regions of the domain. The calculation of these annual values is based on the sum of the seasonal fluxes. Model results reveal that, regionally, the *NEM* supports about 50% of the estuarine CO_2 outgassing, while nitrification and riverine DIC inputs sustain about 17% and 33% of the CO_2 emissions, respectively. The relative significance of the three processes described above shows important spatial variability. In the NAR, oversaturated riverine waters and NEM respectively sustain 50% and 44% of the outgassing within the sub-region, while nitrification is of minor importance (6%). In the MAR, the contribution of riverine DIC inputs is significantly lower (~30%) and the main contribution to the outgassing is *NEM* (~50%); nitrification accounting for slightly less than 20% of the outgassing. In the SAR, the riverine contribution is even lower (~20%), and the outgassing is mainly attributed to the *NEM* (~55%) and nitrification (~25%). Therefore, although the model results reveal significant variability across individual systems, a clear latitudinal trend in the contribution to the total FCO_2 emerge from the analysis; the importance of oversaturated riverine water decreasing from North to South, while *NEM* and nitrification increase along the same latitudinal gradient. The increasing relative importance of

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estuarine biogeochemical processes over riverine DIC inputs as drivers of FCO_2 along the North-South gradient is largely driven by increasing temperatures from North to South, especially in the SAR region (Tab. SI 1).

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Contrasting patterns across the 3 regions can also be observed with respect to carbon filtering capacities, *CFilt* (Fig. 10b). In the NAR, over 90% of the riverine carbon flux is exported to the coastal ocean. However, in the MAR, the high efficiency of the largest systems in processing organic carbon results in a regional *CFilt* that exceeds 50%. This contrast between the NAR and the MAR and its potential implication for the carbon dynamics of the adjacent continental shelf waters has already been discussed by Laruelle et al. (2015). In the NAR, short estuarine residence results in a much lower removal of riverine carbon by degassing compared to the MAR. Laruelle et al. (2015) suggested that this process could contribute to the weaker continental shelf carbon sink adjacent to the NAR, compared to the MAR. In the SAR, most estuaries remove between 40% and 65% of the carbon inputs. The high temperatures observed and resulting accelerated biogeochemical process rates in this region favor the degradation of organic matter and contribute to increase the estuarine filtering capacity for carbon. However, in the SAR, a large fraction of the OC loads is derived from adjacent salt marshes located along the estuarine salinity gradients, thereby reducing the overall residence time of OC within the systems. The filtering capacity of the riverine OC alone, which transits through the entire estuary, would thus be higher than the one calculated here. As a consequence, highest C retention rates are expected in warm tidal estuaries devoid of salt marshes or mangroves (Cai, 2011).

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3.2 Seasonal variability of estuarine carbon dynamics

Carbon dynamics in estuaries of the US East coast not only show a marked spatial variability, but also vary on the seasonal timescale. Table 5 presents the seasonal distribution of *NEM* and FCO_2 for each sub-region. In the NAR, a strong seasonality is simulated for the *NEM* and the summer period contributes more than a third to the annually integrated value. The outgassing reveals a lower seasonal variability and is only slightly higher than summer outgassing during fall and lower during spring. In the MAR, summer contributes more to the *NEM* (>28% of the yearly total) than any other season, but seasonality is less pronounced than in the NAR. Here, FCO_2 is largest in winter and particularly low during summer. In the SAR, summer accounts for 30 % of the NEM, while spring contributes 21 %. FCO_2 is relatively constant throughout the year suggesting that seasonal variations in carbon processing decrease towards the lower latitudes in the SAR. This is partly related to the low variability in river discharge throughout the year in lower latitudes (Tab. S1). In riverine dominated systems with low residence times, such as, for instance, the Altamaha River estuary, the CO_2 exchange at the air-water interface is mainly controlled by the river discharge because the time required to degrade the entire riverine organic matter flux exceeds the transit time of OC through the estuary. Therefore, the riverine sustained outgassing is highest during the spring peak discharge periods. In contrast, the seasonal variability in FCO_2 in long-residence, marine-dominated systems with large marsh areas (e.g. Sapelo and Doboy Sound) is essentially controlled by seasonal temperature variations. Its maximum is reached during summer when marsh plants are dying and decomposing, as opposed to spring when marshes are in their productive stage (Jiang et al., 2008). These contrasting seasonal trends have already been reported for different estuarine systems in Georgia, such as the Altamaha Sound, the Sapelo Sound and the Doboy Sound (Cai, 2011). At the scale of the entire East coast of the US, the seasonal trends in *NEM* reveal a clear maximum in summer and minimal values during autumn and winter. The seasonality of FCO_2 is much less pronounced because the outgassing of oversaturated riverine waters throughout the year contributes to a large fraction of the FCO_2 and dampens the effect of the temperature dependent processes (*NEM* and denitrification). In our simulations, the competition between temperature and river discharge is the main driver of the seasonal estuarine carbon dynamics is. When discharge increases, the carbon loads increase proportionally and the residence time within the system decreases, consequently limiting an efficient degradation of organic carbon input fluxes. In warm

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regions like the SAR, the temperature is sufficiently high all year round to sustain high C processing rates and this explains the reduced seasonal variability in NEM.

3.3 Regional carbon budget: a comparative analysis

The annual carbon budget for the entire East coast of the US is summarized in Fig. 11a. The total carbon input to estuaries along the East coast of the US is 4.6 Tg C y^{-1}, of which 42% arrives in organic form and 58% in inorganic form**.** Of this total input, saltmarshes contribute 0.6 Tg C yr^{-1}, which corresponds to about 14% of the total carbon loads and 32% of the organic loads in the region. The relative contribution of the saltmarshes to the total carbon input increases towards low latitudes and is as high as 60% in the SAR region. Model results suggest that 2.7 Tg C y^{-1} is exported to the continental shelf (25% as TOC and 75% as DIC), while 1.9 Tg C y^{-1} is emitted to the atmosphere. The overall carbon filtering capacity of the region thus equals 41% of the total carbon entering the 43 estuarine systems (river + saltmarshes). Because of the current lack of a benthic module in C-GEM, the water column carbon removal occurs entirely in the form of CO_2 outgassing and does not account for the potential contribution of carbon burial in sediments. The estimated estuarine carbon retention presented here is thus likely a lower bound estimate. Reported to the modeled surface area of the region, the total FCO_2 of 1.9 Tg C y^{-1} translates into a mean air water CO_2 flux of about 14 mol C m^{-2} y^{-1}. This value is slightly higher than the estimate of 10.8 mol C m^{-2} y^{-1} calculated by Laruelle et al., (2013) on the basis of local $\overline{FCO_2}$ estimates assumed to be representative of yearly averaged conditions (see section 2.1). The latter was calculated as the average of 13 annual $\overline{FCO_2}$ values reported in the literature (Tab. 2), irrespective of the size of the systems. This approach is useful and widely used to derive regional and global carbon budgets (Borges et al., 2005; Laruelle et al., 2010; Chen et al., 2013). However, it may lead to potentially significant errors (Volta et al., 2016a) due to the uncertainty introduced by the spatial interpolation

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of local measurements to large regional surface areas, while useful and widely used to derive regional and global carbon budgets.

Regional C budgets are sparse. To our knowledge, the only other published regional assessment of the estuarine carbon and CO_2 dynamics comes from a relatively well studied region: the estuaries flowing into the North Sea in Western Europe (Fig. 11b). This budget was calculated using a similar approach (Volta 2016a) and thus provides an ideal opportunity for a comparative assessment of C cycling in these regions. However, it is important to note that there are also important differences in the applied model approaches and those differences should be taken into account when comparing the derived budgets. In particular, the NW European study is based on a simulation of the 6 largest systems only (Elbe, Scheldt, Thames, Ems, Humber and Weser), accounting for about 40% for the riverine carbon loads of the region. It assumes that the intensity of carbon processing and evasion in all other smaller estuaries discharging into the North Sea (16 % of the carbon loads) can be represented by the average of the 6 largest system simulation results. In addition, the Rhine-Meuse system, which alone accounts for 44% of the carbon riverine inputs of the region, was treated as a passive conduit with respect to carbon due to its very short freshwater residence time (Abril et al., 2002). The contribution of saltmarshes to the regional carbon budget was also ignored because their total surface area is much smaller than along the US East coast (Regnier et al., 2013b). Another important difference is the inclusion of seasonality in the present study while the budget calculated for the North Sea is derived from yearly average conditions (Volta et al., 2016a).

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Overall, although both regions receive similar amounts of C from rivers (4.6 Tg C y^{-1} and 5.9 Tg C y^{-1} for the East coast of the US and the North Sea, respectively), they reveal significantly different C filtering capacities. While the estuaries of the East coast of the US filter 41% of the riverine TC loads, those from the North Sea only remove 8% of the terrestrial-derived material. This is partly due to the large amounts of carbon transiting through the 'passive' Rhine-Meuse system. The regional filtering capacity is higher (15%) when this system is excluded from the analysis. However, even when neglecting this system, significant differences in filtering efficiencies between both regions remain. FCO_2 from the North Sea estuaries (0.5 Tg C y^{-1}) is significantly lower than the 1.9 Tg C y^{-1} computed for the East coast of the US. The reason for the lower evasion rate in NW European estuaries is essentially twofold. First, the total cumulative surface area available for gas exchange is significantly lower along the North Sea, in spite of comparable flux densities calculated using the entire estuarine surface areas of both regions (14 mol C m^{-2} y^{-1} and 23 mol C m^{-2} y^{-1} for the East coast of the US and the North Sea, respectively). Second, although the overall riverine carbon loads are comparable in both regions (Fig. 11), the ratio of organic to inorganic matter input is much lower in the North Sea area because of the regional lithology is dominated by carbonate rocks and mixed sediments that contain carbonates (Dürr et al., 2005; Hartmann et al., 2012). As a consequence, TOC represents less than 20% of the riverine loads and only 10% of the carbon exported to the North Sea. In both regions, however, the increase of the inorganic to organic carbon ratio between input and output is sustained by a negative NEM (Fig. 11). Although the ratios themselves may significantly vary from a region of the world to the other as evidenced by these two studies, a NEM driven increase of the inorganic fraction within carbon load along the estuarine axis is consistent with the global estuarine carbon budget proposed by Bauer et al. (2013). In the East coast of the US, the respiration of riverine OC within the estuarine filter is partly compensated by OC inputs from marshes and mangroves in such a way that the input and export IC/OC ratios are closer than in the North Sea region.

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3.4 Scope of applicability and model limitations

Complex multidimensional models are now increasingly applied to quantitatively explore carbon and nutrient dynamics along the land-ocean transition zone over seasonal and even annual timescales (Garnier et al., 2001; Arndt et al., 2007, 2009; Arndt and Regnier, 2007; Mateus et al., 2012). However, the application of such complex models remains limited to individual, well-constrained systems due their high data requirements and computational demand resulting from the need to resolve important physical, biogeochemical and geological processes on relevant temporal and spatial scales. The one-dimensional, computationally efficient model C-GEM has been specifically designed to reduce data requirements and computational demand and to enable regional/global scale applications (Volta et al., 2014, 2016a). However, such a low data demand and computational efficiency inevitably requires simplification. The following paragraphs critically discus these simplifications and their implications.

Spatial resolution

Here, C-GEM is used with a 0.5° spatial resolution. While this resolution captures the features of large systems, it is still very coarse for relatively small watershed, such as those of the St. Francis River, Piscataqua River, May River or the Sapelo River. For instance, the 5 estuaries reported by Hunt et al. (2010, 2011, see section 2.6) are all small systems contained by the same watershed at a 0.5° resolution. Only watersheds whose area spans several grid cells can be properly identified and represented (i.e. Merrimack or Penobscot with 6 and 9 cells, respectively).

Hydrodynamic and Transport Model

C-GEM is based on a theoretical framework that uses idealized geometries and significantly reduces data requirements. These idealized geometries are fully described by three, easily obtainable geometrical parameters (B, b_0, H). The model thus approximates the variability of estuarine width and cross-section along the longitudinal axis through a set of exponential functions. A comprehensive sensitivity study (Volta et al., 2014) has shown that integrated process rates are generally sensitive to changes in these geometrical parameters because of their control on estuarine residence times. For instance, Volta et al. (2014) demonstrated that the NEM, is particularly sensitive to the convergence length. Similarly, the use of constant depth profile may lead to variations of about 10% in NEM (Volta et al., 2014). Nevertheless, geometrical parameters are generally easy to constrain, especially well-monitored regions such as the US east coast. Here, all geometrical parameters are constrained on the basis of observed estuarine surface areas and average water depths. In addition, the model also accounts for the slope of the estuarine channel. This approach ensures that simulated estuarine surface areas, volumes and, thus, residence times are in good agreement with those of the real systems and minimizes uncertainties associated to the physical set-up.

In addition, the one-dimensional representation of the idealized estuarine systems does not resolve two- or three-dimensional circulation features induced by complex topography and density driven circulation. While C-GEM performs well in representing the dominant longitudional gradients, its applicability to branched systems or those with aspect ratios for which a dominant axis is difficult to identify (e.g. Blackwater estuary, UK; Pearl River estuary, China; Tagus estuary, Portugal; Bay of Brest, France) is limited.

Biogeochemical Model

Although the reaction network of C-GEM accounts for all processes that control estuarine FCO_2 (Borges and Abril, 2012; Cai, 2011), several, potentially important processes, such as benthic-pelagic exchange processes, phosphorous sorption/desorption and mineral precipitation, a more complex representation of the local phytoplankton community, grazing by higher trophic levels, or multiple reactive organic carbon pools are not included. Although these processes are difficult to constrain and their importance for FCO_2 is uncertain, the lack of their explicit representations induces uncertainties in Cfilt. In particular, the exclusion of benthic processes such as organic matter degradation and burial in estuarine sediments could result in an underestimation of Cfilt. However, because very little is known on the long term fate of organic carbon in estuarine sediments, setting up and calibrating a benthic module proves a difficult task. Furthermore, to a certain degree model parameters (such as organic matter degradation and denitrification rate constant) implicitly account for benthic dynamics. We nonetheless acknowledge that, by ignoring benthic processes and burial in particular, our estimates for the estuarine carbon filtering may be underestimated, particularly in the shallow systems of the SAR.

Biogeochemical model parameters for regional and global applications are notoriously difficult to constrain (Volta et al., 2016b). Model parameters implicitly account for processes that are not explicitly resolved and their transferability between systems is thus limited. In addition, published parameter values are generally biased towards temperate regions in industrialized countries (Volta et al., 2016b). A first order estimation of the parameter uncertainty associated to the estuarine carbon removal efficiency (CFilt) can be extrapolated from the extensive parameter sensitivity analyses carried out by Volta et al. (2014, 2016b). These comprehensive sensitivity studies on end-member systems have shown that the relative variation in Cfilt when a number of key biogeochemical parameters are varied by two orders of magnitude varies by is ±15 % in prismatic (short residence time on order of days) to ±25 % in funnel-shaped (long residence time) systems. Thus, assuming that uncertainty increases linearly between those bounds as a function of residence time, an uncertainty estimate can be obtained for each of our modelled estuary. With this simple method, the simulated regional Cfilt of 1.9 Tg C yr-1 would be associated with an uncertainty range comprised between 1.5 and 2.2 Tg C yr^{-1}. Our regional estuarine CO_2 evasion estimate is thus reported with moderate confidence. Furthermore, in the future, this uncertainty range could be further constrained using statistical methods such as Monte Carlo simulations (e.g. Lauerwald et al., 2015).

Boundary Conditions and Forcings

In addition, simulations are only performed for climatological means over the period 1990-2010 without resolving interannual and secular variability. Boundary conditions and forcings are critical as they place the modelled system in its environmental context and drive transient dynamics. However, for regional applications, temporally resolved boundary conditions and forcings are difficult to constrain. C-GEM places the lower boundary condition 20 km from the estuarine mouth into the coastal ocean and the influence of this boundary condition on simulated biogeochemical dynamics is thus limited. At the lower boundary condition, direct observations for nutrients and oxygen are extracted from databases such as the World Ocean Atlas (Antonov et al., 2014). However, lower boundary conditions for OC and pCO_2 (zero concentration for OC and assumption of pCO_2 equilibrium at the sea side) are simplified. This approach does not allow addressing the additional complexity introduced by biogeochemical dynamics in the estuarine plume (see Arndt et al., 2011). Yet, these dynamics only play a secondary role in the presented study that focuses on the role of the estuarine transition zone in processing terrestrial-derived carbon.

Constraining upper boundary conditions and forcings is thus more critical. Here, C-GEM is forced by seasonally-averaged conditions for Q, T, and radiation. To date, GlobalNEWS only provide yearly-averaged conditions for a number of upper boundary conditions (Seitzinger et al., 2005; Mayorga et al., 2010), representative of the year 2000. Simulations are thus only partly transient (induced by seasonality in Q, T and radiation) and do not resolve short-lived events such as storms or extreme drought conditions. In addition, direct observations of upper boundary conditions are rarely available- in particular over seasonal or annual timescales. For the US East Coast estuaries, direct observations are only available for O_2, chlorophyll a, DIC and Alk. For DIC and alkalinity and boundary conditions are constrained by calculating the average concentration over a period of about three decades. In addition, observational data is extracted at the station closest to the model's upper boundary, which might be still located several kilometres upstream or downstream of the model boundary. Upper boundary conditions of POC, DOC, DIN, DIP, DSi are extracted from GlobalNews and thus model-derived. As a consequence, our results are thus intimately dependent on the robustness of the GlobalNEWS predictions. These values are usually only considered robust estimates for watersheds larger than ~10 cells (Beusen et al., 2005), which only correspond to 13 of the 43 estuaries modelled in this study.

Model-data comparison

The generic nature of the applied model approach and, in particular the application of seasonally/annually averaged or model-deduced boundary conditions renders a direct validation of model results on the basis of local and instantaneous observational data (e.g. longitudinal profiles), which is likely not representative of these long-term average conditions, difficult. Therefore, model performance is evaluated on the basis of spatially aggregated estimates (e.g. regional FCO_2 estimates based on local measurements) rather than system-to-system comparisons with longitudinal profile from specific days. However, note that the performance of C-GEM has been intensively tested by specific model-data comparisons for a number of different systems (e.g. Volta et al., 2014, 2016a) and we are thus confident of its predictive capabilities.

Despite the numerous simplifying assumptions inevitably required for such a regional assessment of carbon fluxes along the land-ocean continuum, the presented approach does nevertheless provide an important step forward in evaluating the role of land-ocean transition systems in the global carbon cycle. It provides a first robust estimate of carbon dynamics based on a theoretically well-founded and carefully tested, spatially and temporally resolved model approach. This approach provides novel insights that go beyond those gained through traditionally applied zero-salinity method or box model approaches. In addition, it also highlights critical variables and data gaps and thus helps guide efficient monitoring strategies.

3.5 Towards predictors of the estuarine carbon processing

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The mutual dependence between geometry and transport in tidal estuaries and, ultimately, their biogeochemical functioning (Savenije, 1992; Volta et al., 2014) allows relating easily extractable parameters linked to their shape or their hydraulic properties to biogeochemical indicators. In this section, we explore the relationships between such simple physical parameters and indicators of the estuarine carbon processing $\overline{NEM}$, $\overline{FCO_2}$ and *CFilt*. In order to account for the effect of temperature on C dynamics, -$\overline{NEM}$ and $\overline{FCO_2}$ are also normalized to the same temperature (arbitrarily chosen to be 0 degree). These normalized values are obtained by dividing -$\overline{NEM}$ and $\overline{FCO_2}$ by a Q_{10} function *f(T)* (see Volta et al., 2014). This procedure allows accounting for the exponential increase in the rate of several temperature dependent processes contributing to the NEM (i.e. photosynthesis, organic

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carbon degradation…). Applying the same normalization to -$\overline{NEM}$ and $\overline{FCO_2}$ is a way of testing how intimately linked *NEM* and FCO_2 are in estuarine systems. Indeed linear relationships relating one to the other have been reported (Mayer and Eyre, 2012). The three indicators are then investigated as a function of the ratio between the estuarine surface *S* and the seasonal river discharge *Q*. The surface area is calculated from the estuarine width and length, as described by equation 2, in order to use a parameter which is potentially applicable to other regions for which direct estimates of the real estuarine surface area is not available. Since the fresh water residence time of a system is obtained by dividing volume by river discharge, the S/Q ratio is also intimately linked to residence time. Here, we choose to exclude the estuarine depth from the analysis because this variable cannot be easily quantified from maps or remote sensing images and would thus compromise the applicability of a predictive relationship on the global scale. However, from dimensional analysis, S/Q can be viewed as a water residence time normalized to meter depth of water. As shown by equation 3, *S* only requires constraining *B0* and width convergence length *b*, two parameters that can readily be extracted from the Google Earth engine. Global database of river discharges, as for instance RivDIS (Vörösmarty et al., 1996) are also available in such a way that the S/Q ratio can potentially be extracted for all estuaries around the globe.

Figure 12a reveals that small values of S/Q are associated with the most negative $\overline{NEM}$ / $f(T)$. The magnitude of the $\overline{NEM}$ then exponentially decreases with increasing values of S/Q. Estuaries characterized by small values of *S/Q* are mainly located in the NAR sub-region and correspond to small surface area, and thus short residence time systems. It is possible to quantitatively relate -$\overline{NEM}$ $/f(T)$ and S/Q through a power law function ($y = 25.85\ x^{-0.64}$ with a $r^2 = 0.82$). The coefficient of determination remains the same when excluding estuaries from the NAR region and the equation itself is not significantly different, although those estuaries on their own do not display any statistically significant trend (Tab. 6). The decrease in the intensity of the net ecosystem metabolism in larger estuaries (Fig 8), characterized by high S/Q ratios, can be related to the extensive

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consumption of the organic matter pool during its transit through the estuarine filter. However, when reported to the entire surface area of the estuary, larger systems (with high values of S/Q) still reveal the most negative surface integrated *NEM* (Fig. 12b). It can also be noted that some estuaries from the NAR region display very low values of –*NEM*. These data points correspond to fall and winter simulations for which the temperature was relatively cold (<5 °C) and biogeochemical processing was very low.

The overall response of $\overline{FCO_2}/f(T)$ to S/Q is comparable to that of -$\overline{NEM}/f(T)$ (Fig. 12c), with lower values of $\overline{FCO_2}$ observed for high values of S/Q. However, for S/Q < 3 days m^{-1}, the $\overline{FCO_2}$ values are very heterogeneous and contain many, low $\overline{FCO_2}$ outliers from the NAR region. These data points generally correspond to low water temperature conditions which keep pCO_2 low, even if the system generates enough CO_2 internally via NEM. Thus, the well-documented correlation between $\overline{NEM}$ and $\overline{FCO_2}$ (Maher and Eyre, 2012) does not seem to hold for systems with very short residence times. For systems with S/Q > 3 days m^{-1}, we obtain a regression FCO_2 = -0.64 x *NEM* + 5.96 with a r^2 of 0.46, which compares well with the relation FCO_2 = -0.42 x *NEM* + 12 proposed by Maher and Eyre (2012) who used 24 seasonal estimates from small Australian estuaries. However, our results suggest that this relationship cannot be extrapolated to small systems such as those located in the NAR. Figure 12d, which reports non-normalized FCO_2 reveals a monotonous increase of FCO_2 with S/Q. This suggests that, unlike the *NEM* for which the normalization by a temperature function allowed explaining most of the variability; FCO_2 is mostly controlled by the water residence time within the system. Discharge is the main FCO_2 driver in riverine dominated systems, while interactions with marshes are driving the outgassing in marine dominated systems surrounded by marshes. Net aquatic biological production (NEM being negative or near 0) in large estuaries (with large S/Q) is another important reason for low FCO_2 in such systems. For example, despite the higher CO_2 degassing flux in the upper estuary of the Delaware, strong biological CO_2 uptake in the mid-bay and near zero NEM in the lower bay result in a much lower FCO_2 for the entire estuary (Joesoef et al.

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2015). In systems with S/Q < 3 days m^{-1}, the short residence time prevents the excess CO_2 of oversaturated water from being entirely exchanged with the atmosphere and simulations reveal that the estuarine waters are still oversaturated in CO_2 at the estuarine mouth. Thus, the inorganic carbon, produced by the decomposition of organic matter, is not outgassed within the estuary but exported to the adjacent continental shelf waters. This result is consistent with the observation-based hypothesis of Laruelle et al. (2015) for the NAR estuaries. As a consequence of the distinct behavior of short residence time systems, the coefficient of determination of the best-fitted power law function relating $\overline{FCO_2}$ and S/Q is only significant if NAR systems are excluded ($y = 31.64\ x^{-0.58}$ with a $r^2 = 0.70$). This thus suggest that such relationships (as well as that proposed by Maher and Eyre, 2012) cannot be applied to any system but only those for which S/Q>3 day m^{-1}.

Finally, Fig. 12e reports the simulated mean seasonal carbon filtering capacities as a function of the depth normalized residence time. Not surprisingly, and in overall agreement with previous studies on nutrient dynamics in estuaries (Nixon et al., 1996), the carbon filtering capacity increases with S/Q. The best statistical relation between *CFilt* and S/Q is obtained when including all 3 regions, resulting in $r^2 = 0.70$ ($y = 40.64 \log_{10}(x) + 11.84$). Very little C removal occurs in systems with S/Q < 1 day m^{-1}. For systems characterized by longer depth-normalized residence times, *CFilt* increases regularly, and reaches 100% for S/Q > 100 day m^{-1}. Such high values are only observed for very large estuaries from the MAR region (Delaware and Chesapeake Bays); the majority of our systems had an *S/Q* range between 1 and 100 day m^{-1}. The quantitative assessment of estuarine filtering capacities is further complicated by the complex interplay of estuarine and coastal processes. Episodically, marked spatial variability in concentration gradients near the estuarine mouth may lead to a reversal of net material fluxes from coastal waters into the estuary (Regnier at al., 1998; Arndt et al. 2011). Our results show that this feature is particularly significant for estuaries with a large width at the mouth and short convergence length (funnel shaped or 'Bay type' systems). These coastal nutrient and carbon inputs influence the internal estuarine C dynamics and lead to filtering capacities that

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can exceed 100%. This feature is particularly significant in summer, when riverine inputs are low and the marine material is intensively processed inside the estuary.

Previous work investigated the relationship between fresh water residence time and nutrient retention (Nixon et al., 1996; Arndt et al., 2011; Laruelle, 2009). These studies, however, were constrained by the scarcity of data. For instance, the pioneering work of Nixon et al. (1996) only relied on a very limited number (<10) of quite heterogeneous coastal systems, all located along the North Atlantic. Here, our modeling approach allows us to generate 172 (43 x 4) data points, each representing a system-scale biogeochemical behavior. Together, this database spans the entire spectrum of estuarine settings and climatic conditions found along the East coast of the US. In addition, the ratio S/Q used as master variable for predicting temperature normalized -$\overline{NEM}$, $\overline{FCO_2}$ and *CFilt* only requires a few easily accessible geometric parameters (B0, b and L) and an estimate of the river discharge. While it is difficult to accurately predict $\overline{FCO_2}$ for small systems such as those located in the NAR region, the relationships found are quite robust for systems in which S/Q > 3 days m^{-1}. Most interestingly, *CFilt* values reveal a significant correlation with S/Q and could be used in combination with global riverine carbon delivery estimates such as GlobalNews 2 (Mayorga et al., 2010) to constrain the estuarine CO_2 evasion and the carbon export to the coastal ocean at the continental and global scales.

4. Conclusions

This study presents the first complete estuarine carbon budget for the East coast of the US using a modeling approach. The structure of the model C-GEM relies on a restricted number of readily available global datasets to constrain boundary conditions and limits the number of geometrical and physical parameters to be constrained. Our simulations predict a total CO_2 outgassing of 1.9 Tg C y^{-1} for all tidal estuaries of the East coast of the US. This quantification accounts for the seasonality in estuarine carbon processing as well as for distinct individual behaviors among estuarine types (marine or river dominated). The total carbon output to the coastal ocean is estimated at 2.7 TgC y^{-1}, and the carbon filtering capacity with respect to riverine, marshes and mangrove inputs is thus on the order of 40%. This value is significantly higher than the recently estimated C filtering capacity for estuaries surrounding the North Sea using a similar approach (Volta et al., 2016a), mainly because the surface area available for gas exchange and the draining lithology limits the CO_2 evasion in the NW European systems. At the regional scale of the US East coast estuaries, net heterotrophy is the main driver (50%) of the CO_2 outgassing, followed by the ventilation of riverine supersaturated waters entering the estuarine systems (32%) and nitrification (18%). The dominant mechanisms for the gas exchange and the resulting carbon filtering capacities nevertheless reveal a clear latitudinal pattern, which reflects the shapes of estuarine systems, climatic conditions and dominant land-use characteristics.

Our model results are used to derive predictive relationships relating the intensity of the area-based Net Ecosystem Metabolism ($\overline{NEM}$), air-water CO_2 exchange ($\overline{FCO_2}$) and the carbon filtering capacity (*CFilt*) to the depth normalized residence time, expressed as the ratio of the estuarine surface area to the river discharge. In the future, such simple relationships relying on readily available geometric and hydraulic parameters could be used to quantify carbon processing in areas of the world devoid of direct measurements. However, it is important to note that such simple relationships are only valid over the range of boundary conditions and forcings explored and may not be applicable to conditions that fall outside of this range. In regions with better data coverage, such as the one investigated here, our study highlights that the regional-scale quantification, attribution, and projection of estuarine biogeochemical cycling are now at reach.

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5. Acknowledgements

G. G. Laruelle is Chargé de recherches du F.R.S.-FNRS at the Université Libre de Bruxelles. The research leading to these results has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 643052

(C-CASCADES project). The authors thank V. L. Mulder for her thorough reading of the manuscript upon submission.

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Table 1: Estimates of total annual riverine input from watersheds to estuaries (Tg C yr^{-1}). The ranges are based on Stets and Striegl (2012), Global NEWS (Mayorga et al. 2010), Hartmann et al. (2009), SPARROW (Shih et al. 2010) and DLEM (Tian et al. 2010, 2012). Modified from Najjar et al. 2012.

	DIC	**DOC**	**POC**	**TOTAL**
NAR	0.2-0.8	0.3-2.1	0.1-0.2	0.6-3.1
MAR	1.4-1.8	0.5-2.3	0.1-0.3	2.0-4.4
SAR	0.4-1.4	0.9-1.6	0.1-0.2	1.4-3.2
TOTAL	2.0-4.0	1.7-6.0	0.3-0.7	4.0-10.7

Table 2: Published local annually averaged estimates of $\overline{FCO_2}$ in mol C m^{-2} yr^{-1} for estuaries along the East coast of the US."

Name	**Lon**	**Lat**	**$\overline{FCO_2}$ Observed.**	**Modeled**	**Reference**
Altamaha Sound	-81.3	31.3	32.4	72.7	Jiang et al. (2008)
Bellamy	-70.9	43.2	3.6	3.9	Hunt et al. (2010)
Cocheco	-70.9	43.2	3.1	3.9	Hunt et al. (2010)
Doboy Sound	-81.3	31.4	13.9	25.7	Jiang et al. (2008)
Great Bay	-70.9	43.1	3.6	3.9	Hunt et al. (2011)
Little Bay	-70.9	43.1	2.4	3.9	Hunt et al. (2011)
Oyster Bay	-70.9	43.1	4	3.9	Hunt et al. (2011)
Parker River estuary	-70.8	42.8	1.1	3.9	Raymond and Hopkinson (2003)
Sapelo Sound	-81.3	31.6	13.5	20.6	Jiang et al. (2008)
Satilla River	-81.5	31	42.5	25.7	Cai and Wang (1998)
York River	-76.4	37.2	6.2	8.1	Raymond et al. (2000)
Hudson River	-74	40.6	13.5	15.5	Raymond et al. (1997)
Florida Bay	-80.68	24.96	1.4	n.a.	Dufore (2012)

Table 3: State variables and processes explicitly implemented in CGEM.

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State variables		
Name	**Symbol**	**Unit**
Suspended Particulate Mater	SPM	gL^{-1}
Total Organic Carbon	TOC	µM C
Nitrate	NO_3	µM N
Ammonium	NH_4	µM N
Phosphate	DIP	µM P
Dissolved Oxygen	DO	µM O_2
Phytoplankton	Phy	µM C
Dissolved Silica	dSi	µM Si
Dissolved Inorganic Carbon	DIC	µM C
Biogeochemical reactions		
Name	**Symbol**	**Unit**
Gross primary production	GPP	µM C s^{-1}
Net primary production	NPP	µM C s^{-1}
Phytoplankton mortality	M	µM C s^{-1}
Aerobic degradation	R	µM C s^{-1}
Denitrification	D	µM C s^{-1}
Nitrification	N	µM N s^{-1}
O_2 exchange with the atmosphere	FO_2	µM O_2 s^{-1}
CO_2 exchange with the atmosphere	FCO_2	µM C s^{-1}
SPM erosion	E_{SPM}	gL^{-1} s^{-1}
SPM deposition	D_{SPM}	gL^{-1} s^{-1}

Table 4: Yearly averaged surface area (*S*), fresh water discharge (*Q*), residence time (*Rt*), FCO_2 and *NEM* of all simulated estuaries.

long *degrees*	lat *degrees*	*S* *km²*	*Q* *m³s⁻¹*	*Rt* *days*	$\overline{FCO_2}$ *mol C m⁻² yr⁻¹*	$\overline{NEM}$ *mol C m⁻² yr⁻¹*	FCO_2 *10⁶ mol C yr⁻¹*	*NEM* *10⁶ mol C yr⁻¹*
NAR								
-67.25	44.75	7	38.5	15	3.7	-37.4	27	-270
-67.25	45.25	12	73.6	15	6.0	-56.7	71	-666
-67.25	45.25	12	73.6	15	13.8	-56.6	162	-666
-67.75	44.75	3	68.5	4	6.7	-63.5	23	-221
-68.25	44.75	14	69.5	19	4.1	-56.2	58	-791
-68.75	44.75	89	309.9	23	27.4	-58.2	2431	-5163
-69.75	44.25	50	626.6	5	32.3	-74.4	1607	-3703
-70.25	43.75	3	25.8	10	2.1	-21.0	7	-71
-70.75	41.75	288	103.6	958	5.0	-4.0	1428	-1146
-70.75	42.25	63	210.7	40	16.2	-32.9	1025	-2081
-70.75	42.75	17	105.8	3	56.3	-69.0	943	-1155
MAR								
-70.75	43.25	31	29.9	11	21.6	-37.4	662	-1146
-71.25	41.75	257	28.2	808	3.9	-2.5	997	-650
-71.75	41.25	21	112.4	4	35.2	-32.6	726	-672
-72.75	40.75	20	25.4	62	30.7	-21.1	623	-430
-72.75	41.25	10	142.5	2	150.8	-36.9	1578	-386
-72.75	41.75	55	476.6	3	55.9	-45.7	3088	-2523
-73.25	40.75	19	26.8	56	31.4	-28.4	608	-550
-74.25	40.75	1192	608.2	126	15.5	-11.8	18432	-14047
-75.25	38.25	399	80.5	172	13.9	-5.0	5558	-2016
-75.25	38.75	354	31.8	357	7.5	-3.0	2659	-1076
-75.25	39.75	1716	499.0	221	10.0	-7.8	17072	-13439
-75.75	39.25	224	18.3	434	7.5	-2.9	1685	-640
-76.25	39.25	3427	717.1	352	8.1	-5.1	27646	-17352
-76.75	37.25	586	272.3	74	15.0	-10.4	8810	-6084
-76.75	37.75	154	36.3	163	10.7	-6.6	1654	-1023
-76.75	39.25	59	71.2	29	48.6	-34.6	2862	-2038
-77.25	38.25	206	30.2	268	6.1	-3.3	1265	-676
-77.25	38.75	568	259.2	118	16.7	-10.8	9488	-6134
SAR								
-78.25	34.25	48	167.4	7	122.5	-62.4	5916	-3015
-79.25	33.25	47	56.3	42	43.4	-36.5	2056	-1728
-79.25	33.75	45	291.4	8	85.1	-78.7	3843	-3551
-79.75	33.25	25	33.8	15	37.9	-32.8	956	-828
-80.25	32.75	25	31.0	50	48.8	-42.5	1214	-1057
-80.25	33.25	92	75.5	61	62.7	-61.2	5769	-5625
-80.75	32.25	71	21.1	182	12.9	-7.0	918	-501
-80.75	32.75	164	63.1	95	20.6	-11.5	3372	-1879
-81.25	31.75	92	71.7	45	25.7	-20.9	2361	-1926
-81.25	32.25	130	379.8	11	51.7	-39.2	6732	-5097
-81.75	30.75	34	18.7	61	17.5	-14.7	602	-505
-81.75	31.25	130	17.7	294	5.5	-4.0	713	-523
-81.75	31.75	56	350.5	4	72.7	-67.4	4068	-3770

Deleted: **Table 3**: Published local annually averaged estimates of $\overline{FCO_2}$ for estuaries along the East coast of the US. ¶
Name

Table 5: Seasonal contribution to FCO_2 and *NEM* in each the sub-region. The seasons displaying the highest percentages are indicated in bold. Winter is defined as January, February and March, Spring as April, May and June and so on…

Region	*NEM*	winter	spring	summer	fall	FCO_2	winter	spring	summer	fall
	mol C y^{-1}	%	%	%	%	mol C y^{-1}	%	%	%	%
NAR	-16.3 10^9	14.7	21.2	**37.0**	27.2	7.2 10^9	26.3	18.9	26.5	**28.3**
MAR	-72.2 10^9	21.9	25.9	**28.8**	23.4	108.3 10^9	**29.8**	23.3	20.7	26.2
SAR	-30.5 10^9	24.6	20.9	**30.3**	24.2	39.2 10^9	26	23.4	**27**	23.6

Table 6: Regressions and associated coefficient of determination between the depth normalized residence time (S/Q) and -$\overline{NEM}$ / *f(T)*, $\overline{FCO_2}$ / *f(T)* and *CFilt*.

Region	$-\overline{NEM}/f(T)$	$\overline{FCO_2}/f(T)$	*CFilt*
NAR	$y = 27.84\ x^{-0.17}$	$y = 6.07\ x^{0.00}$	$y = 15.08\ \log_{10}(x) + 4.86$
	$r^2 = 0.11$	$r^2 = 0.00$	$r^2 = 0.40$
MAR	$y = 26.03\ x^{-0.63}$	$y = 34.36\ x^{-0.58}$	$y = 40.46\ \log_{10}(x) + 9.60$
	$r^2 = 0.86$	$r^2 = 0.68$	$r^2 = 0.70$
SAR	$y = 28.36\ x^{-0.71}$	$y = 32.82\ x^{-0.66}$	$y = 23.19\ \log_{10}(x) + 43.71$
	$r^2 = 0.76$	$r^2 = 0.80$	$r^2 = 0.46$
MAR + SAR	$y = 25.85\ x^{-0.64}$	$y = 31.64\ x^{-0.58}$	$y = 33.30\ \log_{10}(x) + 24.88$
	$r^2 = 0.82$	$r^2 = 0.70$	$r^2 = 0.57$
NAR + MAR + SAR	$y = 28.98\ x^{-0.66}$	$y = 12.98\ x^{-0.33}$	$y = 40.64\ \log_{10}(x) + 11.84$
	$r^2 = 0.82$	$r^2 = 0.30$	$r^2 = 0.70$

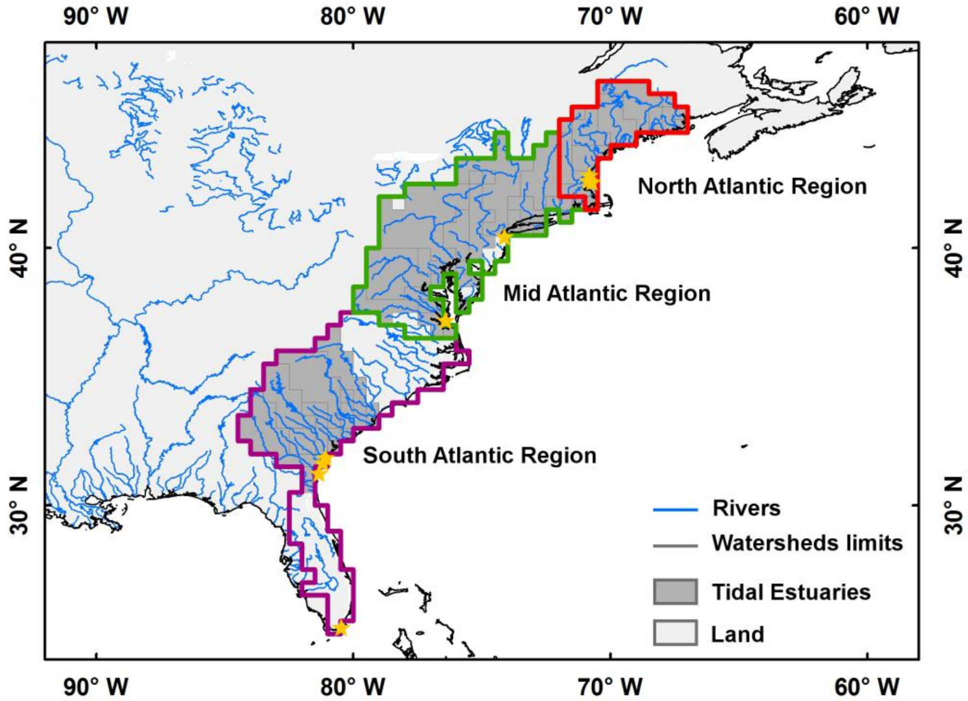

Figure 1: Limits of the 0.5 degrees resolution watersheds corresponding to tidal estuaries of the East coast of the US. 3 sub-regions are delimited with colors and orange stars represent the location of previous studies.

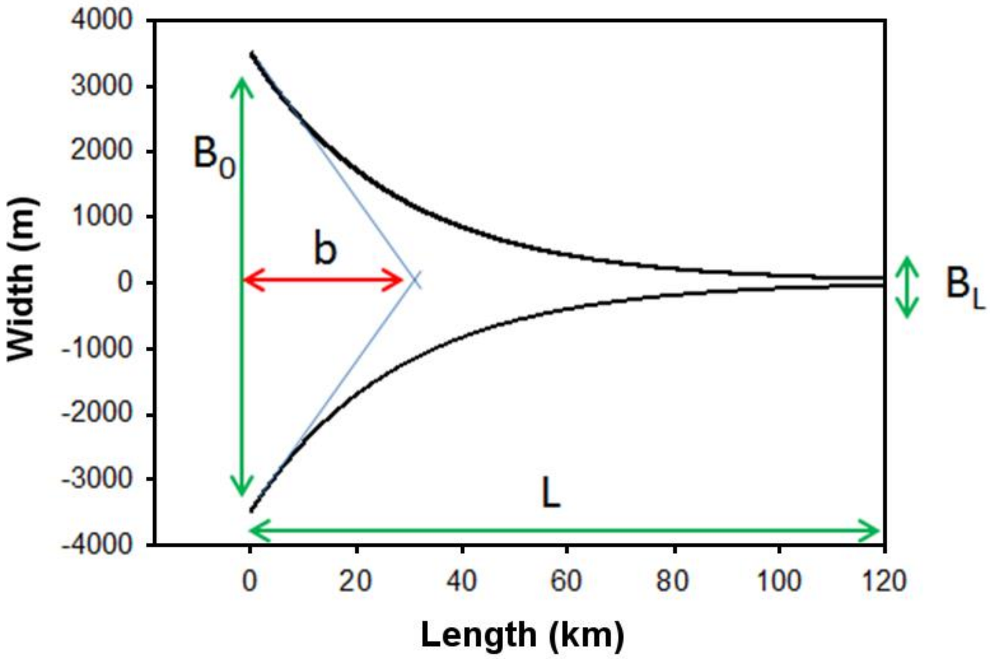

Figure 2: Idealized estuarine geometry and main parameters. Parameters indicated by green arrows are measured, b is calculated. See section 2.3.1 for further details.

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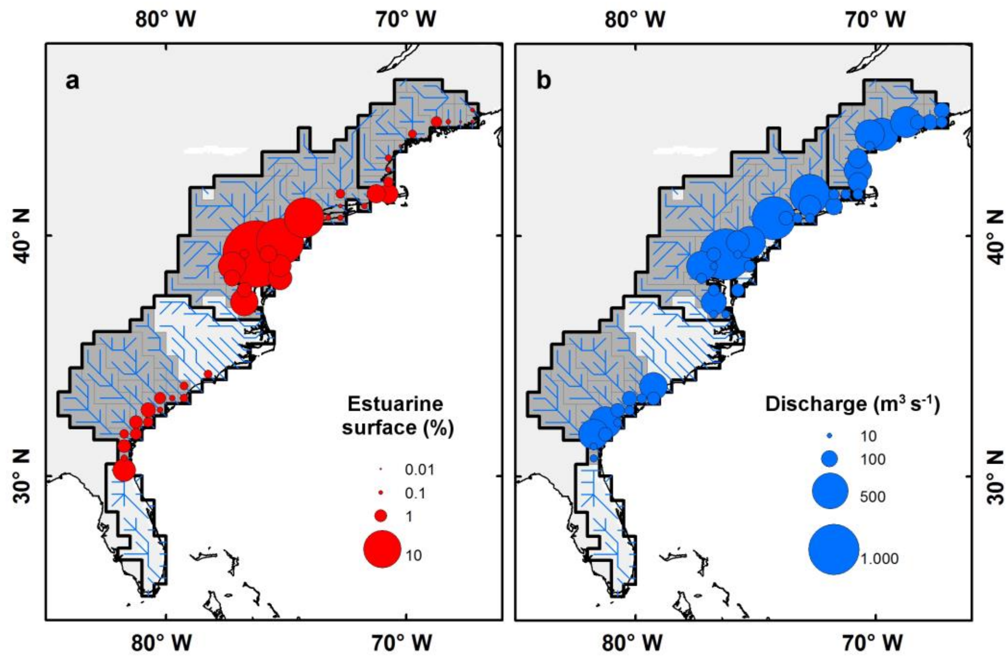

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1750 **Figure 3:** Estuarine surface area (a) and mean annual freshwater discharge (b) for each tidal estuary
1751 of the East coast of the US. Estuarine surface area are expressed as percentage of the entire surface
1752 area of the region (19830 km^2)

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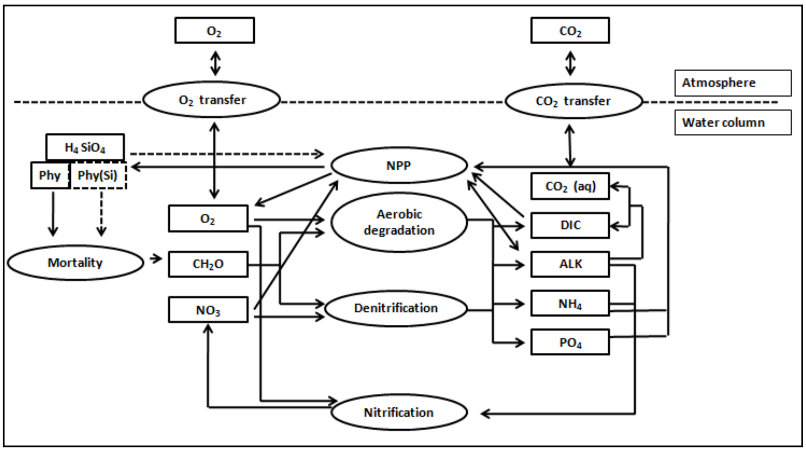

Figure 4: Conceptual scheme of the biogeochemical module of C-GEM used in this study. State-variables and processes are represented by boxes and oval shapes, respectively. Modified from Volta et al., 2014.

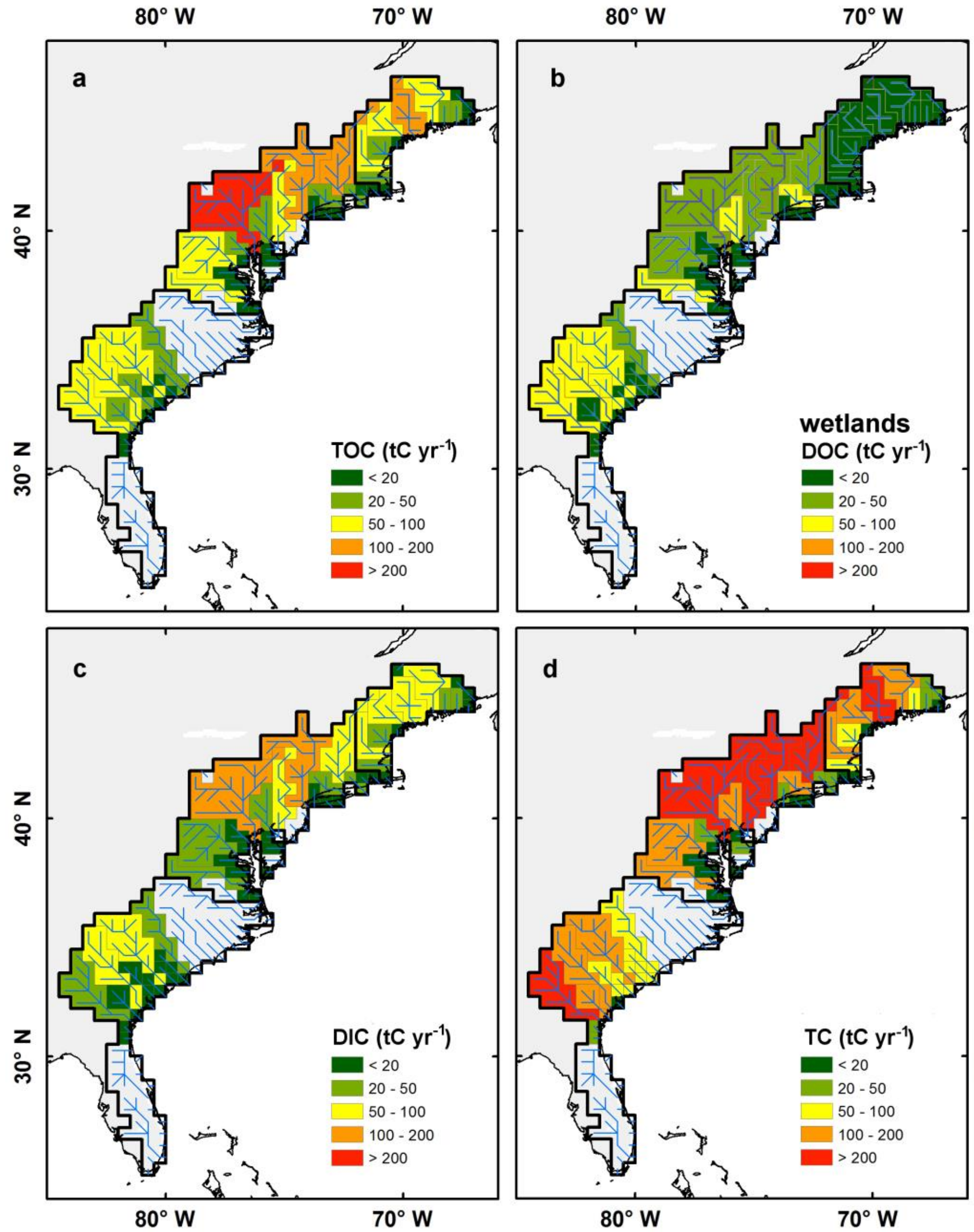

Figure 5: Annual river carbon loads of TOC (a), annual DOC fluxes from wetlands (b), annual river carbon loads of DIC (c) and annual TC fluxes (d). All fluxes are indicated per watershed.

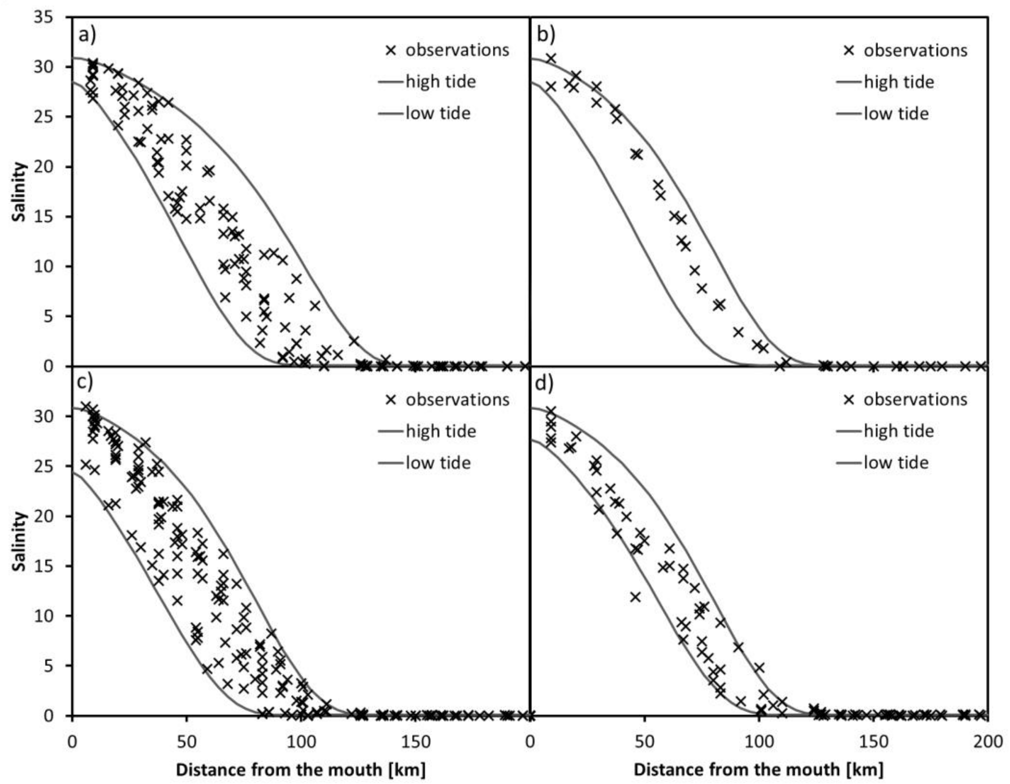

Figure 6. Modeled (lines) and measured (crosses) salinities in the Delaware Bay estuary for January (a), February (b), May (c), June (d). The two lines correspond to high and low tides.

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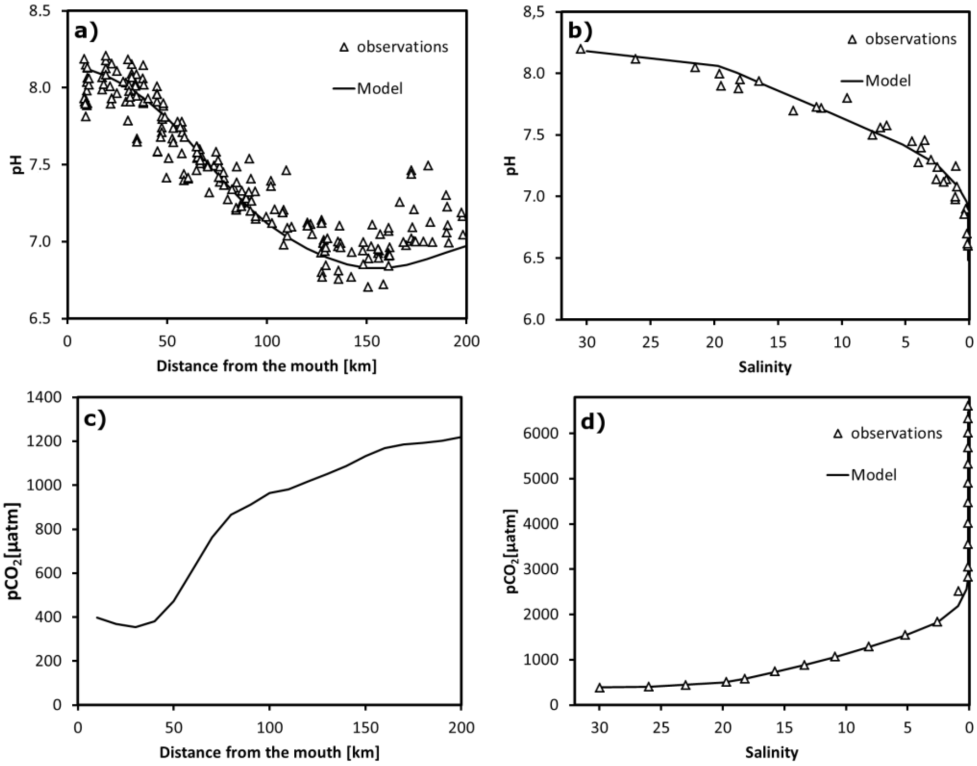

Figure 7. Longitudinal profiles of pH (top) and pCO_2 (bottom) for the Delaware Bay (left) and Altamaha river estuary (right).

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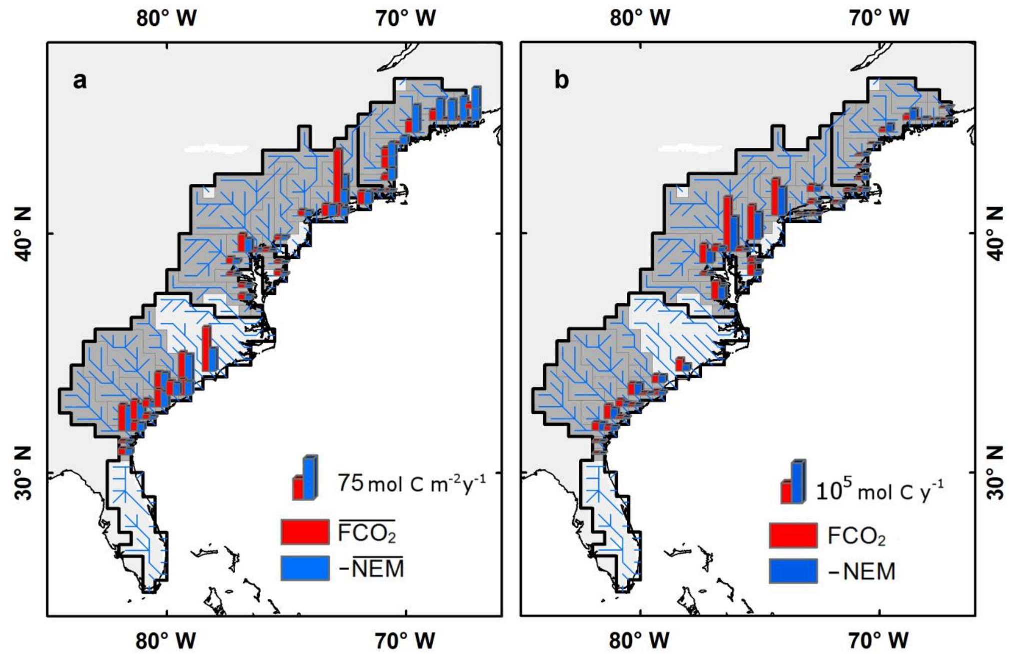

Figure 8: Spatial distribution of spatially averaged value (a) and integrated value (b) of mean annual *FCO_2* (red) and *-NEM* (blue) along the East coast of the US. On panel a, the notation with overbars ($\overline{FCO_2}$ and -$\overline{NEM}$) represents rates per unit surface. For the sake of the comparison with $\overline{FCO_2}$, Fig. 8 displays -$\overline{NEM}$ because the model predicts that all estuaries in this region are net heterotrophic.

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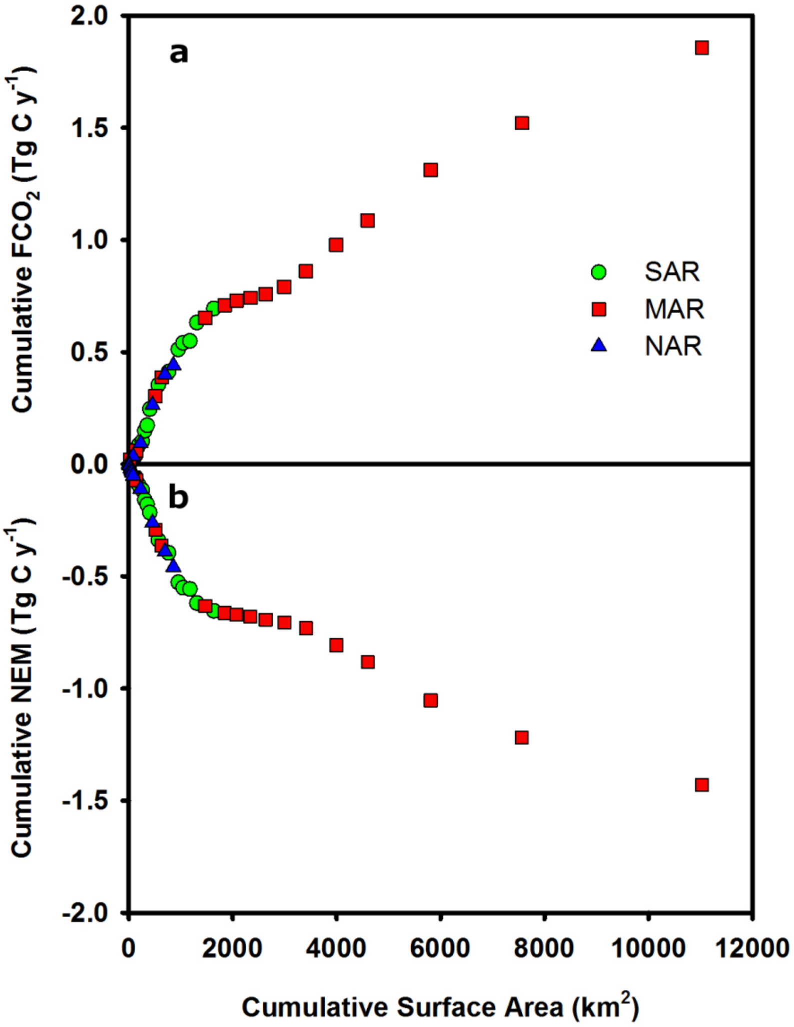

Figure 9: The Cumulative FCO_2 (a) and *NEM* (b) as functions of the cumulative estuarine surface area. Systems are sorted by increasing surface area.

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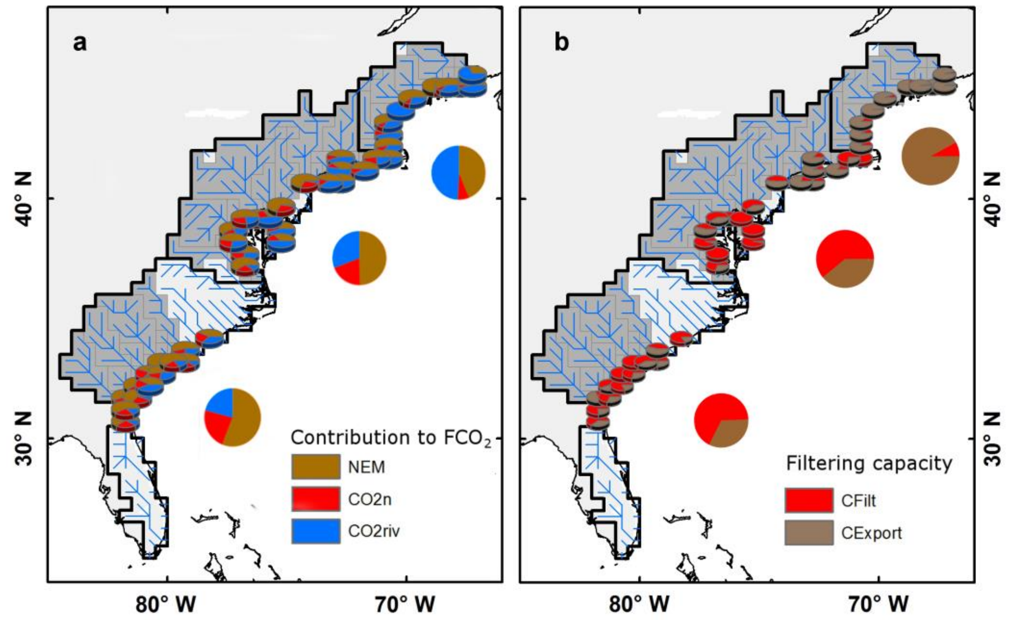

1786

1787 **Figure 10:** Contribution of *NEM*, nitrification and riverine waters super-saturated waters to the mean
1788 annual FCO_2 (a). Spatial distribution of mean annual carbon filtration capacities (*CFilt*) and export
1789 (*CExport*) along the East coast of the US (b).

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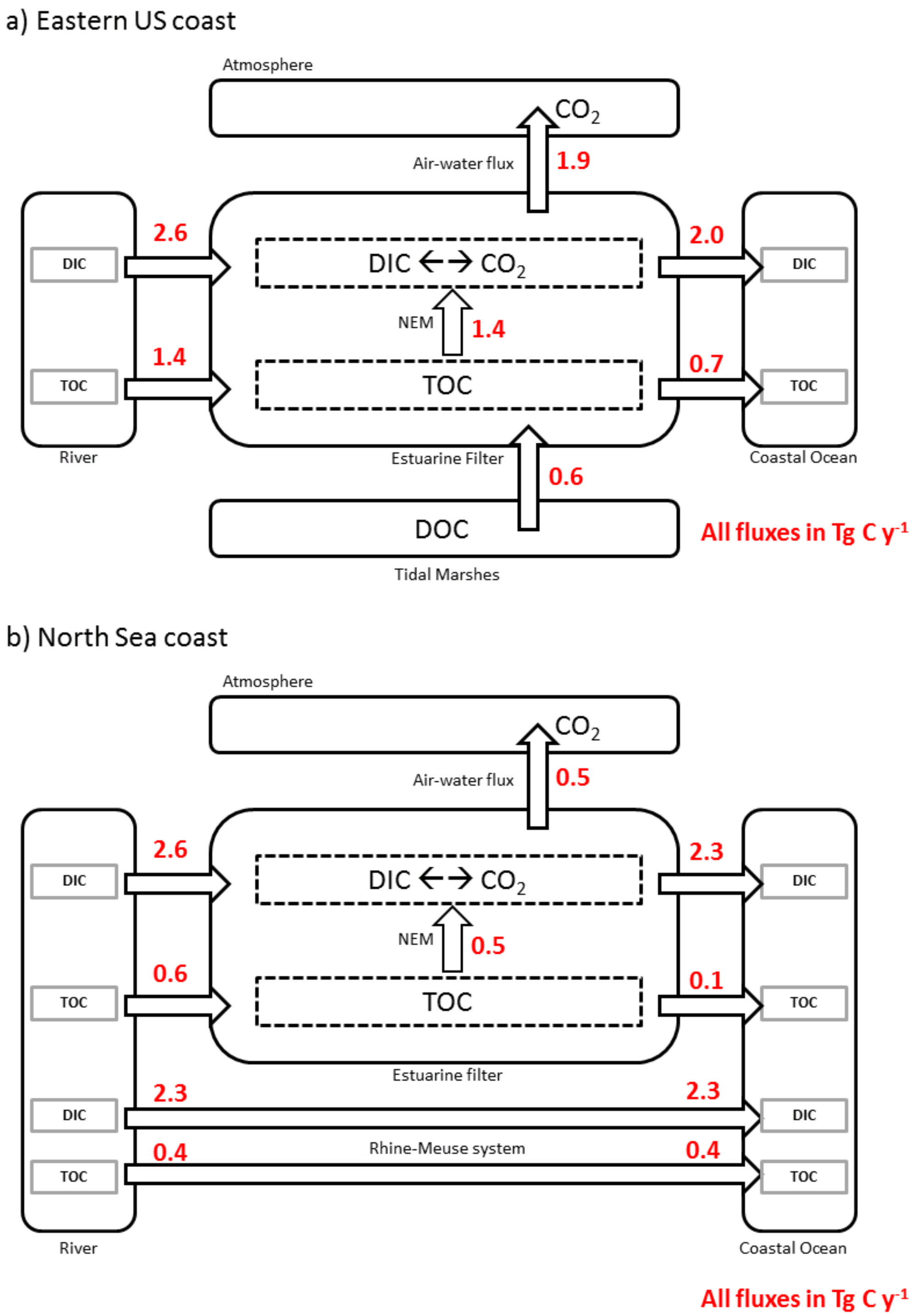

Figure 11: Annual carbon budget of the estuaries of the East coast of the US (a) and of the coast of the North Sea (b, modified from Volta et al., 2016a).

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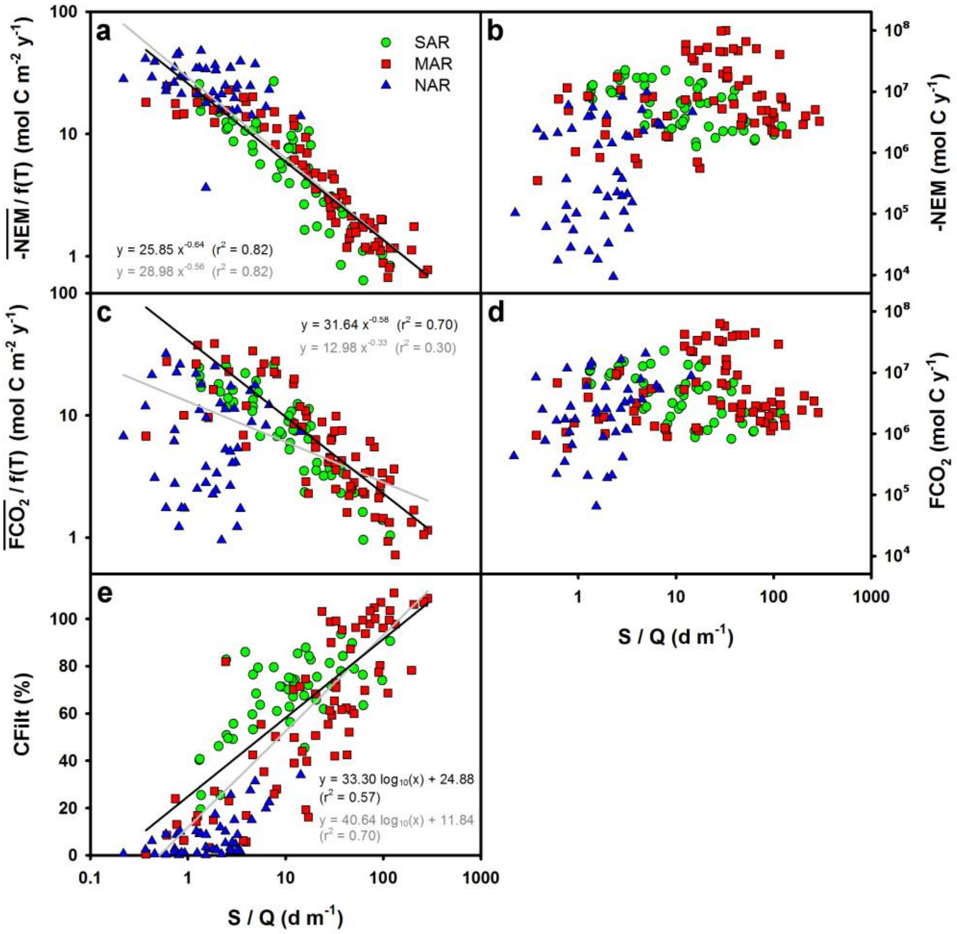

Figure 12: System scale integrated biogeochemical indicators expressed as functions of the depth normalized residence time expressed as the ratio of the estuarine surface S and the river discharge Q for all seasons. Panels b, d and e represent NEM, $-FCO_2$ and CFilt, respectively. Panels a and c represent NEM, $-FCO_2$ normalized by a temperature Q_{10} function. Black lines are the best fitted linear regressions obtained using all the point. Grey lines are best fit using only the estuaries from the MAR and SAR regions.