

# *Interactive comment on* "C-GEM (v 1.0): a new, cost-efficient biogeochemical model for estuaries and its application to a funnel-shaped system" by C. Volta et al.

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Received and published: 16 April 2014

## Response to Referee #2 (Anonymous)

Dear Referee,

We would like to thank you for your constructive comments. Please, find a detailed answer to your comments below. For clarity, our replies are highlighted in blue, while quotes of updated manuscript sections are indicated in red.

C3006

## **General comments:**

The paper by C. Volta et al deals with the modelling of estuarine biogeochemistry and transfer in the context of its complex hydrology. The precise aim of this paper, as quoted in its abstract, is to produce a "generic" model which can be applied to data-poor estuaries, and which aims to be applied on a global scale. Indeed, this is a need of the global carbon community to be able to quantify in a rigorous manner the estuarine filter and the exchange of CO2 in estuaries and deltas Overall, it is a fair attempt to produce such a generic model but it has still a lot of shortcomings which require attention. As quoted by the authors themselves, the main limitation comes from biogeochemical parameters which are neither known nor predictable for unknown environments: the best example is the mineralization rate constant which needs to be tuned for each estuary and to which NEM is very dependent (this is not a large surprise!).

The reviewer emphasizes a very important point here: the lack of a general, objective framework for model parameterization. Models are always simplifications and as such, do not explicitly account for all factors, which control a given biogeochemical process. As a consequence, model parameters that are generally derived by fitting observations implicitly account for the neglected factors. Their implicit nature complicates their transferability and predictive capability to/in data-poor areas. This problem is thus inherent to every biogeochemical model applied to a data-poor area (e.g. the global scale, the past, the future) and results of these models are thus always associated with uncertainties that arise from the implicit nature of model parameters.

C-GEM addresses this issue by:

a) Advocating sensitivity studies to quantify uncertainties.

Extensive parameter sensitivity studies can help quantify uncertainties. The importance of sensitivity studies for the quantification of uncertainties is emphasized throughout the manuscript.

b) Proposing global parameter compilation to help identify trends and constrain

parameters.

Recent global parameter compilations of, for instance, mineralization rate constants in marine sediments (Arndt et al., 2013) have shown promising regional trends. Therefore, the current lack of a general, objective framework for model parameterization should by no means prevent the development of biogeochemical models and their application on the global scale.

c) Resolving the most important temporal and spatial scales and providing an accurate description of the estuarine hydrodynamics and transport.

Currently, assessments of global coastal carbon and nutrient cycling are often based on box model approaches (e.g. Andersson et al., 2005; Slomp and Van Cappellen, 2007; Laruelle, 2009; Mackenzie et al., 2012), which treat the estuary as a single, vertically and horizontally well-mixed box with steady residual hydrodynamic characteristics. Calibrated parameter values thus do not only implicitly account for neglected biogeochemical factors, but also for the unresolved temporal and spatial scales. C-GEM reduces the problem of parameter transferability as it resolves the most important temporal and spatial scales and provides an accurate description of the estuarine hydrodynamics and transport (Figs. 6-8 in the manuscript).

C-GEM thus represents an important step towards a better quantification of carbon and nutrient fluxes on the regional and global scale.

More complicated is the dependence on the geometry of the estuary which at a global scale may be poorly resolved and introduces large variations in NEM because of residence time.

As stated in the manuscript, the required geometric information is readily available and easily extractable from maps and/or remote sensing images. In fact, one of the key advantages of C-GEM is that it only requires a limited set of readily available and thus well-constrained geometric parameters for a realistic representation of the estuarine hydrodynamics and transport.

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I would propose to accept the paper with substantial revisions in order to: - include the limitations of the model by the need of a global parameter set in the abstract and conclusion - discuss shortly the role of very large rivers (Amazon, Mississippi, Changjiang, Congo) which delivers their loads to the continental shelf directly (does the model apply to these very large rivers which deliver 40% of the freshwater?) apply the model on a different system (prismatic?) for which the residence time is shorter, in order to show the capability of the system

As suggested by the reviewer, we included a short discussion of the limitations of the generic modeling approach that arise from the lack of a general framework for biogeochemical model parameterization in the abstract and conclusions (see reply to Comment #1).

In addition, the role of large rivers and the application of C-GEM to such systems are now emphasized (see reply to Comment #2).

We also agree with the referee that an application of C-GEM to different estuarine geometries is very instructive. However, the main purpose of this manuscript is to provide:

a) a comprehensive and technical description of the model approach and the numerical model to which future users will be able to refer to;

b) a fully documented and complete version of the model source code and an example of a model set-up; as well as,

c) a comprehensive assessment of the model's performance.

Additional applications and detailed evaluations of the model to different systems are beyond the scope of this "GMDD model description paper". We choose a tidallydominated, funnel-shaped estuary for a detailed performance test. These estuarine types are generally characterized by long residence times, strong non-linearities in flow and transport and important departures from steady state that together result in a complex interplay of hydrodynamic and biogeochemical processes on different temporal and spatial scales. River-dominated, prismatic estuaries, in turn, are characterized by a strong residual downstream transport and are, thus, less challenging test cases. However, the interested reader can find preliminary model applications across a range of estuarine geometries in Regnier et al., 2013b (a reference has been added).

## Specific comments:

## Comment #1:

include the limitations of the model by the need of parameters sets (in the abstract and conclusion) It is visible from the sensitivity analysis that the main biogeochemical outputs (Remineralisation, denitrification or nitrification) are very sensitive to the parameters used for the model calculation. In the section on "model limitations" (p 5676 line 16-26), the authors state that the lack of such a database for model parameters for tropical or polar regions may limit the use of this model, and claim for the building of a worldwide database for these parameters. This limitation by the lack of large scale parameter sets should be quoted in the abstract (which should be rewritten completely, see detailed comments) and the conclusion. Possible parts to include in the abstract and conclusion stands on page 5675 line 6-14 or page 5676 line 16-26. This part should be explicit in the paper.

A discussion of the limitations that arise from the lack of a general framework for biogeochemical model parameterization has been added to the abstract, which has been rewritten, and the conclusions.

## NEW ABSTRACT:

"Reactive-transport models (RTMs) are powerful tools to disentangle the complex process interplay that drives estuarine biogeochemical dynamics, to assess the quantitative role of estuaries to global biogeochemical cycles and to predict their response to anthropogenic disturbances (land-use change, climate change and

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water management). Nevertheless, the application of RTMs for a regional or global estimation of estuarine biogeochemical transformations and fluxes is generally compromised by their high computational and data demands. Here, we describe C-GEM (Carbon-Generic Estuary Model), a new one-dimensional, computationally efficient RTM that reduces data-requirements by using a generic, theoretical framework based on the direct relationship between estuarine geometry and hydrodynamics. Despite its efficiency, it provides an accurate description of estuarine hydrodynamics, salt transport and biogeochemistry at the appropriate spatio-temporal scales. We provide a detailed description of the model, as well as a protocol for its set-up. The new model is then applied to the funnel-shaped Scheldt estuary (BE/NL), one of the best-surveyed estuarine systems in the world. Its performance is evaluated through comprehensive model-data and model-model comparison. Model results show that C-GEM captures the dominant features of the biogeochemical cycling in the Scheldt estuary. Longitudinal steady-state profiles of oxygen, ammonium, nitrate and silica are generally in good agreement with measured data. In addition, simulated, system-wide integrated reaction rates of the main pelagic biogeochemical processes are comparable with those obtained using a high-resolution, two-dimensional RTM. A comparison of fully transient simulations results with those of a two-dimensional model shows that the estuarine Net Ecosystem Metabolism (NEM) only differs by about 10%, while system-wide estimates of individual biogeochemical processes never diverge by more than 40%. A sensitivity analysis is carried out to assess the sensitivity of biogeochemical processes to uncertainties in parameter values. Results reveal that the geometric parameters LC (estuarine convergence length) and H (water depth), as well as the rate constant of organic matter degradation (kox) exert an important influence on the biogeochemical functioning of the estuary. The sensitivity results also show that, currently, the most important hurdle towards regional or global scale applications arises from the lack of an objective framework for sediment and biogeochemical process parameterization. They, therefore, emphasize the need for a global compilation of biogeochemical parameter values that can help identify common

trends and possible relationships between parameters and controlling factors, such as climate, catchment characteristics and anthropic pressure."

## PAGE 5677, line 19:

"... NEM, and emphasize the need for a global compilation of estuarine sediment and biogeochemical parameters. In addition, such compilation could help identify trends between parameter values and control factors, such as climate, catchment properties and anthropic pressure, and compensate for the current lack of an objective, global framework for parameterization in data-poor areas. The structure ..."

## PAGE 5677, line 26:

"This, together with the compilation of a global dataset for sediment and biogeochemical parameters, could help in the quantification of estuarine biogeochemical cycles at regional and global scales."

## Comment #2:

discuss shortly the role of very large rivers (Amazon, Mississippi, Changjiang, Congo) which delivers their loads to the continental shelf directly Very large rivers which deliver 40% of the freshwater to the global ocean, and have a very different behavior than smaller rivers as the ones dealt with in this paper (Scheldt for exemple). Indeed, a large share of their load is transferred directly to the continental shelf where plume dilution occurs. Is the model able to cope with this type of very large river? This should be specified in the paper, and, if a global vision is the final goal, how to deal with this type of rivers.

C-GEM is designed to model alluvial estuaries. As mentioned in section 2.1, alluvial estuaries are commonly defined as estuaries that are characterized by movable beds and a measurable influence of freshwater inflow. In the case of very large rivers, most of the biogeochemical processing takes place outside of the limits of the

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river mouth, in an external plume considered as an 'external estuary' (e.g. McKee et al., 2004; Dürr et al., 2011). Such large river systems are limited in number (~40 according to Ericson et al., 2006) and are generally well studied (e.g. Gallo and Vinzon, 2005; Denamiel et al., 2013). Considering their tightly coupled river-coastal shelf dynamics and their high contribution to the global riverine water and nutrient discharge, the use of coupled, multi-dimensional local models that resolve both the riverine and coastal shelf environment seems better suited to study their dynmiacs. In addition, the highest resolution Earth System models (0.5 degrees) could represent some of the very large river systems (e.g. Amazon, Mississipi), while the highest resolution Oceanic Global Circulation Models can successfully model the dynamics of their plume (Bernard et al., 2011). While the application of C-GEM to the inland section of a large river estuary, (in order, for example, to provide the inland boundary conditions to a coastal model), is theoretically possible, its main purpose lies in the study of (inner) estuaries, which account for around 30% of the world's river and nutrient loads (Dürr et al., 2011; Regnier et al., 2013b). The application of C-GEM to the inland section of a large river estuary, however (in order, for example, to provide the inland boundary conditions to a coastal model), is likely possible but may not capture the entirety of the biogeochemical processing along the estuarine gradient. We slightly expanded the definition of alluvial estuaries in section 2.1. Furthermore, a discussion of the scope of applicability is now included in Section 5 "Current model limitations", which is now called "Scope of Applicability and Model Limitations".

# PAGE 5650, line 10:

"Alluvial estuaries are commonly defined as systems that are characterized by a movable bed, consisting of sediments of both marine and terrestrial origin, and a measurable influence of freshwater discharge (Savenije, 2005, 2012). In such estuaries, the amount of water flow entering or leaving the estuarine channel is entirely controlled by the shape of the estuary (Pethick, 1984). In turn, the water movement, driven by tides and freshwater discharge, leads to a redistribution of the unconsolidated

sediments and determines the shape of the estuary. Alluvial estuaries display a wide variety of shapes ranging from funnel-shaped estuaries with a dominant tidal influence to prismatic estuaries with a large fluvial influence. Nevertheless, they bear ..."

## PAGE 5651, line 23:

"... characterize tidally-dominated, funnel-shaped estuaries, while fluvial-dominated prismatic estuaries display high N (>15) and S (>15000) and mixed-type estuaries fall in between these two end-member cases. For instance, estuaries such as the Limpopo estuary (Fig. 2a) have a long convergence length and a dominant fluvial influence and show a longitudinal ..."

## PAGE 5676, line 5:

"5 Scope of Applicability and Model Limitations"

#### PAGE 5676, line 10:

"... comparison. However, our ability to assess the role of the estuarine environment for global biogeochemical cycles and greenhouse gas budgets, as well as their response to ongoing global change requires tools that are computationally efficient and can extrapolate knowledge from well-studied to data-poor systems, while at the same time resolving the most important hydrodynamic and biogeochemical processes and scales. The new one-dimensional model C-GEM proposed here is such a computational tool. It represents a valid compromise between performance and computational efficiency and reduces data-requirements by using an idealized representation of the estuarine geometry. Its scope of applicability covers the entire range of alluvial estuaries, from tidally-dominated systems with a large tidal range and low river discharge to fluvial-dominated systems characterized by significant freshwater input (Regnier et al., 2013b). It can be used to resolve the complex process interplay that drives the estuarine biogeochemical dynamic and to quantify estuarine carbon and nutrient budgets. In addition, the computational efficiency of C-GEM offers

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the possibility to simulate simultaneously the biogeochemical dynamics of a large number of estuaries and the contiguous coastal ocean. Although not considered so far, C-GEM could theoretically be applied to the tidally-influenced, inland sections of very large river systems (e.g. Amazon). The value of such application is however questionable because large rivers contribute disproportionally to the overall land to ocean carbon fluxes and might thus deserve a dedicated model. In addition, their tight estuarine-continental shelf coupling and the importance, as well as, the complex multi-dimensional dynamics of their coastal plumes requires a multi-dimensional model representation. Numerous models have already been developed for these systems (e.g. Gallo and Vinzon, 2005; Denamiel et al., 2013) and in the future, they could be explicitly represented in high-resolution Earth System Models (Bauer et al., 2013). In contrast, for the smaller alluvial estuarine systems, mechanistically rooted upscaling strategies need to be designed to better constrain their roles in the global carbon cycle (Bauer et al., 2013) and C-GEM is a tool of choice in this context. However, ..."

#### Comment #3:

apply the model on a different system (prismatic?) for which the residence time is shorter, in order to show the capability of the system I am surprised that the authors did not provide more application cases for the model as this one is supposed to be able to cover a wide range of estuarine functioning. Especially, they quote that sensitivity analysis would be very different if a prismatic system was chosen (page 5675 line 27 to page 5676 line 4). If possible, it would be good to provide the application of the model on another estuary, as this will strengthened the paper.

As already stated above, the main aim of the manuscript is to provide:

a) a comprehensive and technical description of the model approach and the numerical model to which future users will be able to refer to;

b) a fully documented and complete version of the model source code and an example

of a model set-up; as well as,

c) a comprehensive assessment of the model's performance.

We choose a funnel-shaped estuary, with long residence times, strong non-linearities in flow and transport and important departures from steady state for the performance test. Prismatic estuaries are less challenging test cases because their hydrodynamic processes are, due to the strong influence of river discharge and the resulting short residence times, more important than biogeochemical processes. Since the idealized geometry approach is known to be able to capture the main features of estuarine hydrodynamics and transport (e.g. Savenije, 1992, 2005, 2012), the prismatic estuary would not represent an ideal test case. Furthermore, the further application of the model to different systems is beyond the scope of this "GMDD model description paper". However, the interested reader can find preliminary model applications to different estuarine geometries in Regnier et al., 2013b (the reference has been added in the new Section 5).

## Technical comments:

Abstract: the abstract should be rewritten completely as it is more introduction style than abstract. The abstract should provide major results and conclusions of the paper. It is not the case in the present version, as the present abstract just provides the outline of the paper

The abstract has been modified as suggested. See reply to Comment #1.

Page 5632 line 7: ...showing a HALF-gaussian shaped salt intrusion...

We suppose that the referee is referring to page 5652 - line 7, where longitudinal salinity profiles for the three main estuarine types are described. The sentence has been modified as suggested.

C3016

#### PAGE 5652, line 7:

"... showing a half-gaussian shaped salt intrusion curve (Fig. 2b)."

Page 5632 line 19: global biogeochemical cycles... Please specify if major rivers are included or not in this framework. If not modify the sentence.

See reply to Comment #2.

Page 5655 line 25: redSi is not defined in Table 1

Table 1 provides formulations and stoichiometric equations implemented in C-GEM. Biogeochemical parameters used in the C-GEM application, such as Redfield ratios for silica, nitrogen and phosphorous, and their respective values are defined in Table 4.

Page 5665 line 7: the comparison between actual estuarine shape for Scheldt with theoretical description is hard, because Figure 5 is inappropriate. The authors should put the characteristics of Scheldt estuary on the same graph as the model shape, including depth.

As suggested by the referee, the observed estuarine width and depth have been added in panels b and c of Fig. 5. The caption of the figure was modified accordingly.

Page 5666 line 25: "because of heterotrophic nature of the estuary". Do the authors mean the "human impacted nature"?

While the heterotrophic nature of the Scheldt estuary can be partly attributed to the large anthropogenic load it receives (e.g. Vanderborght et al., 2007), other factors

also play a role. For instance, light limitation for instance strongly reduces NPP and thus also favors net heterotrophy. The sentence has been slightly modified.

PAGE 5666, line 25:

"Because of the large anthropogenic influence on the Scheldt estuary, which favours net heterotrophy, nitrogen ..."

Page 5668 line 1-2: sensitivity analysis: remineralisation coefficient can vary on 1 order of magnitude when different types of organic matter are transferred to the estuary. This should be specified in text.

We agree with the reviewer that degradation rate constant can vary depending on the type of organic matter. Nevertheless, different studies indicate that this parameter can vary over more than one order of magnitude (e.g. Soetaert and Herman, 1995; Schroeder, 1997; Arndt et al., 2013). Moreover, other model parameters, such as for instance those related to SPM dynamics, can also vary over a large range. Hence, although we will refer explicitly to the example of degradation constants as suggested, we prefer to keep the sentence generic.

#### PAGE 5668, line 1:

"... and 18). Although sediment and biogeochemical parameters, such as for instance the rate constant of organic matter degradation (e.g. Arndt et al., 2013), can vary over orders of magnitude, here they are varied arbitrarily over a range of  $\pm$ 50% of their baseline value because our aim is to test ..."

Page 5668 line 7: remove "easily"

The word "easily" has been removed.

C3018

Page 5668 line 19: "while it overestimates the tidal amplitude..." please specify by XX%

A sentence, specifying the mean relative error between observed and simulated tidal amplitude values for both estuarine and tidal river zones has been added.

PAGE 5668, line 20:

"... tidal river. In particular, mean relative differences between observed and simulated tidal amplitudes are smaller than 5% and 22% in the saline estuary and in the tidal river, respectively. Discrepancies between model results and observations are ..."

Page 5669 line 19: Figure 6. Please add the envelope of observed SPM in the estuary to allow comparison with the model simulation.

## We suppose that the referee is referring to Fig. 9, showing the simulated, longitudinal SPM profiles.

In our opinion, a comparison between observed SPM profiles and the tidally averaged, steady-state simulation results (displayed in Fig. 9) is not informative. As shown by the long-term record published by Van Damme et al. (2005, Fig. 14), observed SPM concentrations reveal a very patchy pattern in time and space with large fluctuations between 0 and 600 mg l<sup>-1</sup>, resulting from a complex interplay of factors, such as river discharge, tidal forcings, biological activity and terrestrial input of sediments. SPM measurements are typically recorded at different stages of the tidal cycle and under very different forcing conditions and it is, thus, not possible to directly compare the simulated steady-state SPM profile with observed data.

Nevertheless, a general trend, with high values in the Turbidity Maximum Zone (TMZ) and lower concentrations both upstream and downstream of the TMZ, emerges from long-term observations (see Fig. 14 in Van Damme et al., 2005). The longitudinal SPM profile modelled by C-GEM (Fig. 9) is in agreement with this typical pattern and

we are thus confident that the model is able to reproduce, to a first order, the observed SPM dynamics.

Section 4.2.2 has been reformulated to emphasize the difficulty to compare simulated and observed SPM concentrations.

Note that simulated maximum and minimum SPM concentrations have been added to Fig. 9 and the caption has been modified accordingly.

## PAGE 5669, line 14:

"... Arndt et al., 2007). Although SPM concentrations in the Scheldt estuary show a very patchy pattern in time and space due to their high sensitivity to changes in physical forcing conditions (Van Damme et al., 2005), a typical trend, which relates to three well-defined energy regimes along the longitudinal axis of the estuary, can be identified (e.g. Jay et al., 1990; Dalrymple et al., 1992; Arndt et al., 2007). In the lower estuary, where mechanical energy is almost exclusively provided by the tide, observed SPM concentrations are generally low and range between 0 and 150 mg I<sup>-1</sup> (Van Damme et al., 2005). Moving upstream, channel convergence induces an increase in energy dissipation and the associated intensification in tidal amplitude (e.g. Fig. 6) triggers an increase in SPM concentrations from the mouth to the turbidity maximum zone (TMZ), where maximum value of up to 600 mg l<sup>-1</sup> can be observed (Van Damme et al., 2005). The exact location of the TMZ shifts in response to the tidal excursion and the river discharge and is generally found between km 60 and km 100 (e.g. Wollast and Marijns, 1981; Chen et al., 2005). Beyond of the TMZ, friction progressively reduces the tidal influence (Horrevoets et al., 2004) and energy dissipation becomes progressively controlled by the seaward flux of fluvial energy. At the so-called balance point, where both contributions are of similar but low magnitude, low SPM concentrations are typically observed (0-250 mg l<sup>-1</sup>, Van Damme et al., 2005). Upstream of the balance point, close to the estuarine upper limit, the magnitude of the riverine input flux controls the SPM concentration (Chen et al., 2005). The simulated steady-state longitudinal SPM profile (Fig. 9) is in agreement with

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this general pattern. Direct comparison with an observed SPM profile is however not possible because the simulated steady-state conditions do not reproduce a situation observed in the field. SPM concetrations are stongly controled by local exchange processes with the estuarine bed. Hence, already small changes in the physical forcing, as well as their history, exert a large impact on local SPM concentrations and result in large local fluctuations, rendering a direct comparison of simulation results and the range of observed SPM values little informative. Longitudinal ..."

Page 5671 line 20: "while denitrification depends on nitrate production by nitrification". Is that correct for such high NO3 concentration in river (400 umol/l)? is it model output? Please quote a reference if not or explain better.

The sentence referred to the tight link between biogeochemical processes shown in Fig. 11. While nitrate concentrations are indeed high at the upstream boundary, they can reach limiting conditions in the heterotrophic upper reaches and, in particular, around the oxygen minimum (see Fig. 10). We omitted the cited part of the sentence because the following sentence already explains the link between the O2 minimum, nitrification and denitrification rates.

## PAGE 5671, line 18:

"... addition, Figs. 11b-e show that nitrification, denitrification and aerobic degradation are tightly coupled. For instance, high nitrification rates (Fig. 11e) are supported by the ammonium supplied by high aerobic degradation rates (Fig. 11b). Moreover, during summer ..."

Page 5672 line 21: "C-GEM predicts lower NH4 in the tidal river". This is not clearly visible on Fig. 10 where NH4 is plotted versus distance to mouth. You should explain better why nitrification are so much lower in model simulation all over the year.

The sentence referred to results from the transient simulations, while Fig. 10 displays steady-state results. To better explain discrepancies between reaction rates simulated by C-GEM and the two-dimensional model, Section 4.4.3 has been slightly reformulated.

## PAGE 5672, line 5:

"... results. Whole-estuarine aerobic degradation rates are lower than those obtained with the 2-D model during the first period of the year (day < 60), while differences in NPP rates are more pronounced during the summer months. Moreover, C-GEM simulates lower nitrification and denitrification rates. These discrepancies can be traced back to differences in simulated water-depth, estuarine circulation, residence times and/or turbidity. The idealized geometry provides a highly simplified representation of the complex estuarine bathymetry with deep tidal channels and extensive intertidal mud flats. As a consequence, C-GEM ignores the cross-sectional variability in water depth, circulation and, thus, residence times. For instance, C- GEM underestimates residence times in the upper reaches and, therefore, simulates lower biogeochemical rates. These cross-sectional variabilities in residence time, turbidity and residual circulation exert also an important influence on NPP rates. Two-dimensional simulation results highlight the pronounced differences between NPP rates in tidal channels and intertidal flats (e.g. Arndt and Regnier, 2007), a feature that cannot be resolved by the idealized bathymetry of C-GEM. The simplification of the estuarine bathymetry may thus explain also the observed differences in simulated NPP rates. In addition, C-GEM simulates lower nitrification and denitrification rates but slightly higher aerobic degradation rates during the summer months. These discrepancies ..."

Page 5675 line 6-14: include in abstract

Please, refer to reply to Comment #1.

C3022

Page 5676 line 16-26: important for abstract and conclusion

Please, refer to reply to Comment #1.

Page 5677 line 23-27: you should be more cautious with this conclusive statement and quote the two limitations: 1- you need a global set of biogeochemical parameters 2- you ignore major rivers which have a different functioning.

These two aspects have been addressed by modifying the conclusion. Please, refer to replies to Comments #1 and #2.

Figure 13 should be enlarged as it is much too small. Legends are unreadable whatever the magnification, yet this a key Figure of the paper.

We understand the concern of the referee regarding the readability of figure 13. We re-organised the panels and increased the font size.

NB. Please, note that the upgraded manuscript, including new figures (and captions), has been uploaded as supplement.

# Literature cited in the responses:

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