

Reply to Reviewer#1:

Reviewer: This manuscript is based on data from 10 field expeditions carried along different parts of the lowland Congo river. Measurements of water concentrations of CO₂, CH₄, and N₂O is in focus, but in addition many more variables including temperature, conductivity, pH, turbidity, suspended solids, DOC, POC, cations, dissolved silicate, nitrate, nitrite, ammonia, chlorophyll-a, primary productivity, pelagic respiration, and dissolved O₂, methane oxidation, stable isotopic compositions – ¹³C in CH₄, DIC, POC and ¹⁸O in water. In addition to the 10 specific expeditions, there were continuous or regular multi-year monitoring of selected variables on a few sites. Therefore, the data reported is exceptionally rich for any river and even more impressive given that they regard the Congo river. This in itself bring a very high value to the scientific community and we should be grateful for the author's persistence and long-term dedication for studying this very important river basin.

The patterns emerging from this data is very interesting and the authors bring forward a several aspects under the umbrella of a greenhouse gas study. In my view this study contains much more than just greenhouse gases and a general comment would be to broaden the umbrella and title to better reflect all topics actually covered – also those that are not strictly greenhouse gas-related.

In general I find the text well-written and interesting, but I have some questions and comments outlined below that I think need consideration.

Reply : We thank the reviewer for the positive evaluation of our work and for the detailed and useful comments for improvement. Both reviewers suggested changes of the title of paper to reflect that the paper reports data other than GHGs. We decided to keep the title paper focused on GHGs that are the central topic of the paper, while other data gravitate around this central topic. A general title such as “biogeochemistry of the Congo” would in our opinion lead to a loss of visibility and attractiveness of the paper, as we expect that the main readership of the paper will be from the GHG community. We have nevertheless conceded to remove “overwhelmingly driven by fluvial-wetland connectivity” to accommodate reviewer#2's comment on this part of the title.

Reviewer: General comments:

- The manuscript is long and in some parts a bit hard to follow when it combines many aspects and variables in the same paragraphs. I think all information given is valuable so I am not asking for removing any of them, but if possible to provide a more clear structure and to perhaps shorten the text a bit that would make reading and understanding easier.

Reply: We have added new sub-section titles and we have added a few sentences at the start of the results-discussion section that give the “storyline” of the paper to explain to the reader the structure and rational of the paper. This should help the reading and understanding of the paper.

- In some cases when trying to explain observed patterns/correlations, it seems single cause-effect patterns are suggested, while other alternatives may also be possible but not mentioned as the main explanation. : : and sometimes these other

alternatives are brought up later in the text, separated from the first explanation. This regards e.g. to what extent patterns observed in the river depend on in-river processes or on what is brought to the river from the soils. Clearly the authors have expert awareness about all possible explanations, so this is a request to clarify that several alternative explanations are possible when relevant.

Reply: We have modified the text accordingly.

- I miss method evaluations, sensitivity analyses, reliability checks, and attempts to validate estimates regarding the greenhouse gas emissions. This is very important because the suggested extrapolated emissions of CO₂ and CH₄ are exceptionally large – much larger than reported from any other river worldwide - including the Amazon.

There are many potentially very large sources of uncertainty in the flux estimation and its upscaling that is not mentioned nor analyzed, and single approaches from the literature are simply accepted without critical discussions and testing other alternatives.

More details on the two last general comments are provided in the detailed comments below.

Reply: We have added an error analysis, and we estimated the potential export of C from flooded forest to check if it is compatible with the C emissions. Refer to replies below.

Reviewer: Detailed comments:

Reviewer: P2 L15: Please clarify how % values link to atmospheric concentrations, eg is 0% or 100% in equilibrium with the atmosphere?

Reply: This has been clarified in text, 100% corresponds to atmospheric equilibrium.

Reviewer: P2 L19-24. Are these explanations for the observed patterns the only options? Could patterns not also simply correspond to the relative input from anaerobic soil water (higher such input would correspond to lower O₂ and NO₃⁻ but higher NH₄⁺).

Reply: we agree that denitrification can also occur in soils, and we have modified text to mention “soil or sedimentary denitrification”.

Reviewer: Is the abstract the best place for explanations of correlations if there are several alternatives? Is it not better to simply report the correlations in the abstract and, unless reasons are very clear, leave the discussions on explanations to the discussion section?

Reply: we find the explanations given in the abstract clear and concise. We have the impression the reader might want some words of explanation of the variations above and below atmospheric equilibrium.

Reviewer: P2 L32-33: As in the above comment, I wonder if the proposed direct link between pCO₂, CH₄ and DOC variability via organic matter processing is the only

option. Can transport patterns including the balance between input and output be excluded? It seems reasonable that soil water bringing lots of DOC from surrounding soils could also bring lots of CO₂ and CH₄, and that this could explain correlations even if carbon processing in the river itself was not the reason, which is also supported by the following text in the manuscript. Therefore, I find this sentence emphasizing a link between pCO₂ and CH₄ and in-river DOC confusing and also potentially misleading and I suggest to remove it.

Reply: text mentions “intense processing of organic matter” but does not mention that it is in-stream. We agree that this “processing of organic matter” can occur in soils, and in fact we interpret our patterns of CO₂/CH₄ as resulting from “intense processing of organic matter” in flooded forests rather than in-stream. For clarity, we have modified the sentence that now reads : “The wide range of pCO₂ and CH₄ variations was consistent with the equally wide range of %O₂ (0.3-122.8%) and of dissolved organic carbon (DOC) (1.8-67.8 mg L⁻¹), indicative of generation of these two greenhouse gases from intense processing of organic matter either in *terra firme* soils, wetlands or in-stream.”

Reviewer: P5 L24-26: This sentence claim half of the global wetlands in the tropics and 40% in the northern temperate zone, leaving only 10% for the peatlands in the boreal and subarctic zones. This does not fit with e.g. N. C. Davidson A B D , E. Fluet-Chouinard C and C. M. Finlayson A. 2018. Global extent and distribution of wetlands: trends and issues. Marine and Freshwater Research 69(4) 620-627 <https://doi.org/10.1071/MF17019>., or with several other wetland extent studies. Please check and revise sentence.

Reply: The initial statement was correct in the sense that about 50% of wetlands are located between 33°N and 33°S (sub-tropics and tropics); what we called “northern temperate” probably corresponds to what the reviewer refers to “boreal-subarctic”. In order to avoid confusion related to the boundaries between temperate and boreal regions we have rephrased the sentence that now reads: “About half of the global surface area of wetlands is located in the tropics and sub-tropics (33°N-33°S) and the rest in the Northern Hemisphere (Fluet-Chouinard et al. 2015), and more than half of river surface area is located in the tropics and sub-tropics (Allen and Pavelsky 2018)”. We have also updated the reference by Fluet-Chouinard et al. 2015 who proposed two additional products to the GLWD that we initially cited (Lenher & Döll 2004) although the relative distribution per latitude is very similar. The Davidson et al. (2018) cited by the reviewer uses the data originally given by Fluet-Chouinard et al. (2015), so we preferred to cite the original paper.

Reviewer: P8 L6-14: Please give more details on the equilibrator system to prevent dependence on access to key method papers (not easy to access papers for all readers worldwide). What was the water residence time relative to the gas exchange equilibration time in the equilibrator?

Reply: Text now reads: “The equilibrator consisted of a Plexiglas cylinder (height of 70 cm and internal diameter of 7 cm) filled with glass marbles; pumped water flowed from the top to the bottom of the equilibrator at a rate of about 3 L min⁻¹; water residence time within the equilibrator was about 10 secs, while 99% of equilibration was achieved in less than 120 secs (Frankignoulle et al. 2001). This type of

equilibrator system was shown to be the fastest among commonly used equilibration designs (Santos et al. 2012).”

Reviewer: P10 L12-13: Was linear models best for estimating methane oxidation? Sometimes exponential decay model can work better.

Reply: We agree that both linear and exponential decay models can be used to compute MOX, but we found a linear regression to give a good fit of the data.

Reviewer: P11 L31: The used model was developed for closed systems. Why was not the opensteady state system model tried (e.g. Happell, J. D.; Chanton, J. P.; Showers, W. S. Geochim. Cosmochim. Acta 1994, 58, 4377-4388.)?

Reply: We added the following text to justify the choice of the closed system: “The model we used to compute F_{ox} applies for closed systems, implying that CH_4 is assumed not to be exchanged with surroundings in contrast to open system models. Running river water corresponds to a system that is intermediate between closed and open chemical systems, since it is open to the atmosphere and the sediments, but on the other hand the water parcel can be partly viewed as a closed system being transported downstream with the flow. As such, the water parcel receives a certain amount of CH_4 from sediments and then is transported downstream away from the initial input of CH_4 . We also applied two common open-system models to estimate F_{ox} given by Happell et al. (1994) and by Tyler et al. (1997) that have also been applied in lake systems (Bastviken et al. 2002). However, both open-system models gave F_{ox} values > 1 in many cases (data not shown) since the difference between $\delta^{13}C$ of the CH_4 source and measured $\delta^{13}C$ in dissolved CH_4 was often much higher than expected from the assumed isotopic fractionation (1.02). The same observation ($F_{ox} > 1$) was also reported with open-system models in the lakes studied by Bastviken et al. (2002). Since F_{ox} values > 1 are not conceptually possible, we preferred to use instead the results from the closed-system model, although we acknowledge that flowing waters are in fact intermediary systems between closed and open, and that consequently the computed F_{ox} values are under-estimated.”

Reviewer: P14 L26. Please provide the equations used. Readers from all countries may not easily get access to other papers and can then not adopt or evaluate this study as should be possible.

Reply: Equations were added

Reviewer: P18 L2-18 and elsewhere: Some of the discussions on reasons for observed patterns seem to focus entirely on potential explanatory processes in the river channels, while alternative but not always mentioned explanations could relate more to the balance between different compounds entering the channel from the surrounding soils. Please check for possible alternative explanations to the observed patterns and report all relevant alternatives if not clear that a single explanation is most likely.

Reply: Text was modified accordingly.

Reviewer: P29-34 Section 3.4. The mean concentrations found are very high – at least for CH₄. Mean values reported here are more than 2-fold higher than other recent estimates from the Congo basin by Upstill-Goddard et al 2017 (doi.org/10.5194/bg-14-2267-2017).

Reply: Upstill-Goddard et al (2017) sampled a smaller fraction of the Congo basin (5 major river basins) and in the Western side, while we sampled a much larger number of river sites and on the Eastern side, covering river draining the core of the CCC. This explains why we obtain larger values than the estimates of Upstill-Goddard et al (2017). This is unrelated to differences in methodology, as we inter-calibrated our method with the one of used by Prof. Robert Upstill-Goddard, as reported in Wilson et al. (2018, doi: 10.5194/bg-15-5891-2018), although we admit this was for lower ranges of CH₄.

Reviewer: Further, the extrapolated CO₂ emission is 251 TgC yr⁻¹, i.e. 0.25 PgC per year which is more than 10% of the total global estimate of inland water C emissions (2.1 PgC yr⁻¹; Raymond et al 2013 Nature).

Reply: The Congo river accounts for 3% of the total surface of rivers globally, so it's unsurprising that it should also account to a large amount of the total riverine emissions. The fact that our estimates are high compared to those reported by Raymond et al. (2013) is not surprising since the single "average" pCO₂ value used by Raymond et al (2013) for the Congo (derived from a global pH and alkalinity database) is too low compared to our field data (refer to Fig. S18).

Reviewer: This value is also 3.3-fold higher than the total terrestrial NEE of the Congo basin (77 Tg C yr⁻¹ as reported by the authors). Hence, given that not all this terrestrial NEE will be exported laterally to the river, degraded there and be emitted as CO₂, it means that wetlands in the basin not being a part of the terrestrial or the river network carbon balances, presumably would need to have an NEE of > 200-300 TgC or even more, and it is unclear how reasonable this.

Reply: The point is that the export of DOC and CO₂ from wetlands is much larger than from dry forests so that it is not necessary that wetlands have a NEE above 200-300 TgC as suggested by the reviewer. Please also refer to reply below.

Reviewer: It would be good to discuss this and make a reliability check from a catchment carbon mass balance perspective.

Reply: We have addressed this comment and we have added to the discussion the following text: "(...) the carbon export from flooded forests to riverine waters of the Congo basin can be roughly estimated to 396 TgC yr⁻¹ and is in excess of the integrated FCO₂ (based on the export per surface area of flooded forest of 1,100 gC m⁻² yr⁻¹ reported by Abril et al. (2014) for the Central Amazon and on surface area of flooded forest of the CCC (360,000 km⁻², Bwangoy et al. 2010)"

Reviewer: Another concern with the extrapolated values are that a rigorous self-evaluation of the methods used regarding flux estimates seems to be missing. What

are the main uncertainties? How large are the uncertainties? How robust are the area determinations?

What possible area range is there? How about the k estimation? How reliable is that? There are different models to determine k in rivers. Was several of them tried? Do single k models really work across all stream/river orders? Were there any tidal influence that could have affected k in the most down-stream river sections? What ranges were derived? Were there any real in-situ flux measurement that can be used to validate the k estimates? If not, can k or flux estimates be validated in other ways? How about the observed concentrations? In rivers/streams it is often found that water concentrations are regulated by the emission rates so that the highest water concentrations are found where k values and fluxes are lowest, while concentrations are lower where fluxes are high which keep concentrations at low levels. How was this considered in the upscaling? Please add a careful method evaluation and examine and discuss the flux extrapolation critically.

Reply: we have now added a detailed error analysis of the fluxes that answers most of the above questions. To answer the other questions of reviewer: The Congo river is different from (for instance) the Amazon as it is isolated from the ocean by a series of water falls leading to an abrupt altitude change of nearly 280m over a few km (downstream of Malebo pool). So there is no tidal influence on the river Congo “proper”, but only in the estuary “at sea” that we did not sample. We did carry out a limited number of flux measurements with floating chambers that allow to compute k_{600} values. These numbers are briefly discussed and presented by Borges et al. (2015, doi: 10.1038/NGEO2486) but we think this is not the basis to actually validate the procedure to calculate the k_{600} we used in the upscaling, due to a very restricted number of measurements mainly in high order streams. Text now reads “An error analysis on the GHG flux computation and upscaling was carried out by error propagation of the GHG concentration measurements, the k value estimates, and the estimate of surface areas of river channels to scale the areal fluxes, using a Monte Carlo simulation with 1000 iterations. The uncertainty on the GHG concentrations led to an uncertainty of areal fluxes of $\pm 1.2\%$, $\pm 2.3\%$ and $\pm 7.1\%$ for CO_2 , CH_4 and N_2O , respectively. The uncertainty on k based on tracer experiments is typically $\pm 30.0\%$ (Ulseth et al. 2019). This leads to a cumulated uncertainty of areal fluxes of $\pm 17.6\%$, $\pm 17.9\%$ and $\pm 19.0\%$ for CO_2 , CH_4 and N_2O , respectively. The uncertainty of the river/stream surface areas based on GIS analysis of Allen and Pavelesky (2018) is estimated to $\pm 10\%$ leading to an overall uncertainty of integrated fluxes of $\pm 18.3\%$, $\pm 18.3\%$ and $\pm 19.6\%$ for CO_2 , CH_4 and N_2O , respectively.”

Reviewer: Figure 2. Detailed maps with green borders to the right seem not correctly positioned on the central big map.

Reply: We have redrawn the figure to take correct this, and we thank the reviewer for spotting this.

Reviewer: Figure 7. The outliers indicated as red dots do not seem to deviate so much from the other nearby dots. How much difference did removal of them make and is such a removal needed for some good reasons?

Reply: The inclusion of the outliers in the regression decreases slightly the r^2 and changes imperceptibly the slope; but it does not change the sign of the slope

(positive or negative) and does not change the statistical significance of regression. However, one or two points visually stood out, and this was confirmed with the statistical test. We found this information relevant enough to be included in the figures.

Reviewer: Tab S1-S3 in Spatial Analysis supplement: Is it realistic that the velocity was lowest in the highest slope low order streams? I realize this is the consequence of Equation S2 and that the river cross section dimensions change much more than the slope and drives this pattern: : : but is it realistic and was this validated? One could imagine a positive correlation between velocity and slope and that velocity-slope relationship eventually break down or change character for higher order rivers.

Reply: While it is seemingly unintuitive, flow velocity has long been shown, both empirically and theoretically, to be lowest in low-order streams. As the reviewer alluded to, the "increase of velocity downstream results from the fact that the increase in depth overcompensates for the decrease in slope" (Leopold and Maddock, 1953, <https://pubs.usgs.gov/pp/0252/report.pdf>). To answer any questions a reader might have, we added the following sentence to section S2.2 in the Spatial Analysis Supplement: "Although this produces an unintuitive positive relationship between flow velocity and stream order, there is long-standing empirical evidence that shows that mean flow velocity is lower in low-order streams where hydraulic roughness is greatest (Leopold and Maddock, 1953)."