

Response to reviewer 1 (response in red).

We thank Reviewer 1 for taking the time to review the paper. We agree that additional validation, of pCO₂ would improve the paper, as well as some additional analysis of the causes for the discrepancy between our estimates of riverine FCO₂ vs those of Borges et al (2019), such as gas transfer velocity and stream surface area. We respond in more detail below.

The model validation step is very slim, as authors compare the model outputs of dissolved organic carbon (DOC) to a sub-set of published field data. DOC in tropical rivers is extremely refractory and provides little grasp on aquatic carbon cycling as the most labile DOC fraction is very rapidly mineralized (both in soils and in water). So DOC provides a poor validation of the carbon cycling in the river, and might be considered almost as a passive tracer and provide a rough validation of the hydrological connectivity between soils and rivers. Conversely, a convincing validation of the model would be to compare the model outputs of the dissolved CO₂ concentration (or the corresponding partial pressure of CO₂) with the extensive field data collected by Borges et al. (2019) that are publically available (Borges and Bouillon 2019). While a point by point comparison would not make sense, it could be useful to check if the model captures the overall range of spatial variations along the river and among the different stream sizes. Such validation would be extremely convincing because the CO₂ evasion from the river to the atmosphere (the core topic of the paper) is computed from the dissolved CO₂ concentration (the atmospheric CO₂ is comparatively invariant) and the gas transfer velocity. So if the model does not represent correctly the dissolved CO₂ concentration then this implies that the CO₂ evasion rates are incorrect (as well as any conclusion based on past reconstruction and future projection from the model outputs).

Thank you for these suggestions. We have conducted some analysis (in line with your suggestions) of the monthly average pCO₂ over the decade from 2005-2014, and this shows that we are able to capture well, the differences in pCO₂ along the mainstem (and their seasonal variations) outlined in Borges et al (2009). Note that we analysed the decadal average to try to minimise the influence of interannual variation for a given year.

For example, during highwater (mean of 6 consecutive months of highest flow, 2005-2014) we simulate a mean pCO₂ of 3,373 ppm and 5,007 ppm at Kinsangani and Kinshasha respectively, compared to Borges et al's values of 2,424 ppm and 5,343 ppm during highwater (measured in December 2013).

Similarly, during low flow season (mean of 6 consecutive months of lowest flow, 2005-2014) we simulate a mean pCO₂ of 1,568 ppm and 2,643 ppm at Kinsangani and Kinshasha respectively, compared to Borges et al's values of 1,670 ppm and 2,896 ppm during falling water (June 2014).

Moreover, for the Oubangui river at Bangui we simulate a range (2005-2014) of 402 ppm to 3,195 ppm across months, similar to the seasonal variation of 470 to 3,750 observed by Bouillon et al. (2012) over 2010-2011.

However, our simulations fail to capture the very high mean pCO₂ values (i.e. up to 12,000 ppm) observed in the smallest tributaries of the Congo in Borges et al. (2019). The highest mean riverine (in headwaters- represented by the fast reservoir) pCO₂ values that we simulate for the present day are around 9,000 ppm. In ORCHILEAK, note that for the fast reservoir we assume a full pCO₂ equilibrium with the atmosphere over one full day, which prevents very high pCO₂ values from building.

As discussed in the paper, the lack of accounting for Aquatic Macrophytes is likely to be one reason for our relatively low CO₂ evasion from the river surface, amongst others covered in the discussion. However, as noted in the manuscript we estimate a similar overall CO₂ evasion to Borges et al (2019) once we include the flux from floodplains/ wetlands, which account for 85% of our total CO₂ evasion. Moreover, the majority of this flux comes from the Cuvette Centrale (Fig 6) which suggests that while ORHILEAK fails to attribute the majority of this to small rivers (owing to the coarse resolution of the river network in the model at 0.5 degree ~ 50 km) we nonetheless do capture the source of carbon. In other words, in ORCHILEAK the majority of this carbon evades directly from the floodplain and wetlands of the Cuvette Centrale as opposed to the rivers.

In the revised manuscript we would elaborate further on this pCO₂ validation in the results section, and add 1 or possibly 2 new figures based on these results. We would also modify the discussion to reflect these new insights.

The overall emission of CO₂ from the fluvial component of the Congo Basin (TgC/yr) is based on the product of a CO₂ flux density (mol/m²/yr) and a stream surface area; the areal CO₂ flux is itself computed from the air-water CO₂ concentration gradient, and the gas transfer velocity; the air-water CO₂ concentration gradient in turn is mainly function of the dissolved CO₂ concentration. So there are three quantities that could explain the difference between the 4 estimates of integrated CO₂ emissions discussed in section L-513-517: the CO₂ dissolved concentration, the gas transfer velocity and the stream surface area. The evaluation of the model performance would be much more convincing if each of these three quantities was compared to available estimates.

The only study of the Congo which we know of where k₆₀₀ has been measured (albeit, indirectly) is Borges et al. (2015, Nature) based on measurements FCO₂ and pCO₂. They calculate an average k₆₀₀ for the Congo river of 2.9 m d⁻¹, relatively similar to our riverine value used of 3.5 m d⁻¹, which we discuss in line 519. Note also that the sensitivity analysis performed in Lauerwald et al. (2017) showed that in the physical approach of ORCHILEAK, CO₂ evasion is not very sensitive to the k value, unlike data-driven models. Namely, Lauerwald et al (2017) showed that an increase or decrease of k₆₀₀ for rivers and swamps of 50% only led to 1% and -4% change in total CO₂ evasion, respectively. Rather, ORCHILEAK is sensitive to CO₂ inputs from soils and floodplains, as well as instream production through DOC decomposition.

In terms of surface area, we simulate a mean present day River surface area of 25,900 km² compare to 23,190 km² (3% of 773 000 km² – we assume the 3% is a rounded figure so this is likely larger) in Borges et al. (2019, see section 3.8) or the 26,517 km² quoted in Borges et al (2015, Scientific Reports). Therefore, we think that this can be discounted as a major reason for the discrepancy and we will reiterate this in the revised manuscript.

Final elements of validation and discussion that are missing are the contribution of HCO₃⁻ to the export of dissolved inorganic carbon (DIC) from the river to the ocean. The export of DIC from rivers to the ocean is mainly in the form of HCO₃⁻ including the Congo river (Wang et al. 2013). So ignoring HCO₃⁻ would lead to a very substantial under-estimation of DIC export to the ocean. This should be fairly easy to implement with a weathering model and GIS of lithology. This is of course of interest for the topic of paper as weathering intensity is a function of temperature and precipitation that are used by the authors to study the long-term (1861-2099) trends of aquatic carbon fluxes. Further, there are substantial data-sets of total alkalinity (that mainly corresponds to HCO₃⁻) providing

spatial (Borges et al. 2019) and seasonal (Wang et al. 2013; Bouillon et al. 2012; 2013) patterns of HCO₃⁻ variability. Additionally, dissolved CO₂ is in thermodynamic equilibrium with HCO₃⁻, so it is required to have some grasp on HCO₃⁻ variability to correctly model dissolved CO₂ dynamics, hence, CO₂ emissions to the atmosphere.

We appreciate these comments and suggestions and acknowledge that the lack of accounting for HCO₃⁻ is a limitation of the model. However, we would consider creating and implementing a weathering model outside the scope of our study and therefore propose that as alternative, we add estimates from the literature and discuss these limitations with additional caveats.

We estimate a free dissolved CO₂ export to the coast of 2.4 Tg C yr⁻¹ for the present day, relatively similar to the empirically derived estimate of the total DIC export of 3.3 Tg C yr⁻¹ calculated in Wang et al (2013). According to Wang et al, dissolved CO₂ accounts for the majority (1.9 Tg C yr⁻¹) but the difference between our estimate and theirs is indeed likely to be largely caused by our lack of accounting for the weathering derived flux (HCO₃⁻) which they estimate at 1.4 Tg C yr⁻¹. In summary, despite these limitations the results of Wang et al, suggest that we still capture the majority of the DIC flux.

However, Cai et al. (2008, Continental Shelf Research), suggest that the total DIC flux could potentially be substantially larger at around 13 Tg C yr⁻¹. We will discuss these issues in more detail in the revised manuscript.

SPECIFIC COMMENTS L 24 : It's the increase of air temperature rather than "climate change" in general.

Thanks, will change as suggested

L44 : In this section it's unclear what is meant by "increase" of "net primary productivity" and "storage in tree biomass". A recent study shows that African forests are sinks of carbon on yearly basis, and that the carbon sink is constant in time from the mid-1990's to present (Hubau et al. 2020). So, according to this study, there is no "increase" in NPP as stated but a constant sink. Please clarify.

We will modify these sentences in line with the new findings of Hubau et al. 2020. This paper was published after we submitted and previous literature indicated that there was an increase in tree biomass (Lewis et al., 2009) between 1968- 2007. By tree biomass we mean the biomass and in turn carbon storage (Mg C ha⁻¹ yr⁻¹) of intact forests (as surveyed by the team of Lewis et al). Please also see response to Reviewer 2.

L74 : The tropical region is also a hotspot of aquatic C cycling due to wetland productivity and wetland carbon inputs to rivers (Abril & Borges 2019), in addition to "terrestrial NPP". In the Amazon river a large fraction of fluvial CO₂ emissions to the atmosphere are sustained by wetland inputs (Abril et al. 2014). L74: The fact that the "tropical region is a hotspot area for inland water C cycling" (as stated) was authoritatively demonstrated by seminal papers such as Richey et al. (2002) and Melack et al. (2004), more than a decade before the recent Lauerwald et al. (2015) work. L 78-80: there are (at least) two additional elements of context that could be relevant for the introduction: 1) the Raymond et al. (2013) and the Lauerwald et al. (2015) estimates are in fact based on the same initial data-base of pCO₂ computed from pH and alkalinity (GLORICH) that was extrapolated globally using two different approaches; this illustrates how uncertain these global estimates are, since the resulting values differ by a factor of 3; 2) While the conclusion of Lauerwald et al. (2015) that the majority of CO₂ emissions from rivers comes from the tropics is probably correct, field data (used in

the global extrapolation) is nearly absent in the tropics. For instance for the Congo River there is only one single data entry in the GLORICH data-set. Most of the data used to develop the statistical model of Lauerwald et al. (2015) come from non-tropical areas such as North America and Scandinavia.

We will modify the introduction in line with these suggestions, prioritising empirically derived estimates wherever possible, and add additional context.

L 244: Borges et al. (2019) report discharge data from the mainstem Congo at Kisangani. So there are additional data-sets to validate the model hydrology.

We performed the Hydrology validation before this paper was published but would be happy to validate our simulation against this additional data source, or alternatively could change the wording of this sentence.

L513: It could be useful to put into context how these different fluxes were computed. The Raymond et al. (2013) estimate is based on a single pCO₂ value (apparently from pH and alkalinity measurements in Pool Malebo) that was extrapolated to the whole basin. The comparison of this single value of pCO₂ with the extensive data set reported by Borges et al. (2019) shows that it is unrealistically low (refer to Supplemental Figure 18). Lauerwald et al. (2015) et al. estimate of pCO₂ compares better to the Borges et al. (2019) data-set but still fails to represent the influence of the Cuvette Centrale (refer to Supplemental Figure 18). Please also note that the CO₂ estimate for the Congo reported by Borges et al. (2015) was based on exactly the same stream surface area and gas transfer velocity as those used by Raymond et al. (2013), and also showed that the Raymond et al. (2013) estimate was under-estimated (obviously, since the pCO₂ value is unrealistically low). So there is some clear convergence that the present estimate of CO₂ emission based on ORCHILEAK is under-estimated even if it is coincidentally close to the one reported by Raymond et al. (2013). The actual reasons of the under-estimation need to be explored as suggested in the above Major Comments.

We agree that some additional context would be helpful to the reader, as well as the additional validation (of pCO₂ etc) which we cover in our response to your major comments.

In the discussion of the long-term changes of DOC export, it could be useful to mention that there was no measurable difference in DOC flux from the Oubangui (largest tributary of the Congo) between the 1990's (Coynel et al. 2005) and the 2010's as reported and discussed by Bouillon et al. (2012; 2014).

Thank you for pointing this out and we will add a sentence on this.

References

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