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## Carbon leakage through the terrestrial-aquatic interface: Implications for the anthropogenic CO<sub>2</sub> budget

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#### Abstract

This contribution reviews the role of the terrestrial-aquatic interface in the global carbon cycle. We highlight that carbon leakage through this interface has profound ramifications for the terrestrial carbon balance and for the anthropogenic  $CO_2$  budget. Our budget analysis identifies the need for an integrated process-based quantitative understanding of the terrestrial-aquatic interface that includes its response to anthropogenic perturbations. Complementary observational and modeling efforts in this direction are presented.

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#### 1. Introduction

Since the beginning of the Industrial Era,  $CO_2$  emissions due to fossil fuel combustion and cement production ( $E_{FF}$ ) and land-use change ( $E_{LUC}$ ) have led to a rapid accumulation of the carbon mass in the atmosphere<sup>[1-4]</sup>. For the decade 2003-2012,  $E_{FF}$  and  $E_{LUC}$  have been estimated at 8.6±0.4 PgC yr<sup>-1</sup> and 0.9±0.5 PgC yr<sup>-1</sup>, respectively. Altogether, anthropogenic emissions thus amount to 9.5 PgC yr<sup>-1</sup>. Over the same period, the atmospheric  $CO_2$  concentrations have increased by about 20ppm, which translates in a mean annual growth rate  $G_{ATM}$  of 4.3±0.1 PgC yr<sup>-1</sup><sup>[3]</sup>. This implies that about 45 % of the anthropogenic emissions accumulate in the atmosphere, the

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remainder, about 55 %, being taken up by the land (S\_{LAND}) and ocean (S\_{OCEAN}) 'CO\_2 sinks'.

From mass balance, the following equation constrains the anthropogenic CO<sub>2</sub> budget:

 $E_{FF}+E_{LUC}=G_{ATM}+S_{LAND}+S_{OCEAN}$  (1)

The land sink term  $S_{LAND}$  has traditionally been estimated from the closure of the CO<sub>2</sub> budget, a contribution referred to as the 'residual land sink'. Based on the partitioning of anthropogenic carbon, the net storage on land is:

 $\Delta C_{\text{LAND}} = S_{\text{LAND}} - E_{\text{LUC}} \quad (2)$ 

Equation 1 considers only anthropogenic  $CO_2$  emissions and their distribution among the atmosphere, ocean and land. This analysis thus omits that carbon is continuously displaced along the land-ocean aquatic continuum (LOAC) comprising freshwaters, estuaries and coastal waters. A significant fraction of this lateral carbon flux is a steady state component of the pre-industrial carbon cycle that can be ignored in the above budget analysis, which focuses only on the anthropogenic perturbation. However, the remaining fraction, which corresponds to the change in LOAC carbon fluxes since pre-industrial conditions, is a perturbation that is relevant for the global carbon budget. The inclusion of these lateral fluxes of anthropogenic  $CO_2$  will affect the estimates of  $S_{LAND}$  and  $S_{OCEAN}$  in Eq. (1).

#### 2. The land carbon sink and terrestrial carbon storage

For the past five decades, terrestrial ecosystems have been absorbing 25–30% of anthropogenic CO<sub>2</sub> emissions, with a major contribution from the carbon accumulation in forest biomass and soils<sup>[5,6]</sup>. In the classical view which ignores the LOAC carbon cycle, the land sink results from a disequilibrium between terrestrial NPP and soil respiration plus fire disturbance emissions of carbon, initiated by human-induced changes in environmental conditions (both direct effects such as regrowth after land use change, and indirect biogeochemical effects such as the effects of rising CO<sub>2</sub>, climate change and nitrogen deposition), its persistence being related to the residence time of carbon in the terrestrial biosphere (from a few decades to several centuries). Several processes contribute to SLAND, but their precise quantification, both at the regional and global scales, remain entailed with large uncertainties. In particular, the modeling of soil carbon dynamics is poorly constrained<sup>[7]</sup>, as witnessed by the large discrepancies in estimated present-day stocks (from about 500 and 3000 PgC) and predicted future changes (cumulative change of land carbon storage ignoring LOAC, ranging from -165 PgC to +500 PgC for the 21st century, according to the spread between different scenarios and different coupled carbon-climate models<sup>[8]</sup>). Proposed mechanisms behind S<sub>LAND</sub> include the fertilization effect of CO<sub>2</sub> and nitrogen on vegetation growth and increasing growing season length in high latitudes attributed to climate warming<sup>[6,9]</sup>. Overall, S<sub>LAND</sub> mitigates the anthropogenic increase of atmospheric  $CO_2$  levels, and provides a negative feedback in the climate-carbon cycle system<sup>[10]</sup>

Pan et al.<sup>[5]</sup> have estimated forest carbon sources and sinks that allow to constrain  $\Delta C_{LAND}$  (the contribution from croplands and grasslands to biomass accumulation is much smaller). Consecutive forest inventories indicate a substantial 4.0 PgC yr<sup>-1</sup> sink with 2.9 PgC yr<sup>-1</sup> in biomass, 0.9 PgC yr<sup>-1</sup> in litter and soil, and 0.2 PgC yr<sup>-1</sup> in harvest products. This sink is partially offset by emissions from gross deforestation of tropical forests (2.8 PgC yr<sup>-1</sup>) and the net C increase in forest thus reduces to 1.2 PgC yr<sup>-1</sup>. The partition of this net C increase (1.0 PgC yr<sup>-1</sup> without the accumulation into harvest products) between biomass, litter and soil can be calculated if one takes into account that soil carbon loss represents on average 25% of total gross deforestation emissions<sup>[11]</sup>. This leads to about 0.8 PgC yr<sup>-1</sup> accumulation in biomass and 0.2 PgC yr<sup>-1</sup> accumulation in litter and soil<sup>[11]</sup>. In addition, drainage of peatlands contributes to an estimated carbon loss of about 0.4 PgC yr<sup>-1</sup>. Altogether, this leads to a 'bottom-up' estimate of the net carbon sequestration in terrestrial ecosystems of about 0.8 PgC yr<sup>-1</sup>.

#### 3. A leakage in the land carbon sink

The contribution of the Land-Ocean Aquatic Continuum (LOAC) to the global carbon budget is synthesized in

Regnier et al.<sup>[11]</sup>. In what follows, we summarize the main findings concerning the contribution of the LOAC to the CO<sub>2</sub> anthropogenic perturbation and briefly discuss the implications for the estimates of the net carbon storage on land. The anthropogenic C input from terrestrial ecosystems to freshwaters has been estimated at 1 PgC yr<sup>-1</sup>. This input can be decomposed into four fluxes attributed to soil-derived C, chemical weathering of continental surfaces, sewage and net C fixation in inland waters. Altogether, these four perturbation fluxes imply that only a portion of the anthropogenic CO<sub>2</sub> taken up by land ecosystems (S<sub>LAND</sub>) remain sequestered in soil and biomass pools, as 1 ± 0.5 PgC yr<sup>-1</sup> is exported to the LOAC. This land to hydrosphere carbon flux, F<sub>LH</sub>, of magnitude comparable to E<sub>LUC</sub>, is thus significant for the anthropogenic CO<sub>2</sub> budget.

The exported anthropogenic C accumulates to a large part in sediments of freshwaters, estuaries and coastal seas and this term can thus be viewed as the LOAC sink ( $S_{LOAC}$ , 0.55 ± 0.3 PgC yr<sup>-1</sup>). Another fraction is released back to the atmosphere ( $E_{LOAC}$ , 0.35 ± 0.2 PgC yr<sup>-1</sup>), the magnitude of this flux resulting from the combined effects of CO<sub>2</sub> outgassing in inland waters and CO<sub>2</sub> uptake in the coastal ocean<sup>[11,14]</sup>. Finally, a small fraction of anthropogenic C displaced by the LOAC accumulates in the open ocean (0.1 ± > 0.05 PgC yr<sup>-1</sup>).

The carbon budget following Eq.1 implicitly incorporates the fluxes from the LOAC with  $S_{LAND}$ . However, if Terrestrial Ecosystems and the LOAC are separated in the analysis, the residual sink attributed to terrestrial ecosystems alone is now larger (3.15 ± 0.9 PgC yr<sup>-1</sup>) than the value of 2.8 PgC yr<sup>-1</sup> reported for  $S_{LAND}$  in the classical analysis, because this flux is partially offset by  $E_{LOAC}$ . Most importantly, because anthropogenic CO<sub>2</sub> taken up by land ecosystems is exported to the LOAC, the annual land carbon storage change (1.25 PgC yr<sup>-1</sup>) is notably smaller than the net CO<sub>2</sub> uptake by land ecosystems as calculated by Equation 2 (1.9 PgC yr<sup>-1</sup>), a significant fraction of the displaced carbon (0.65 PgC yr<sup>-1</sup>) being stored in freshwater and coastal sediments (S<sub>LOAC</sub>), and to a lesser extent, in the open ocean. With the LOAC included, we now have:

 $\Delta C_{\text{LAND}} = S_{\text{LAND}} - E_{\text{LUC}} - F_{\text{LH}} \quad (3)$ 

The net biomass and soil sequestration estimate calculated according to Equation 3 is broadly consistent with the 'bottom-up' estimates derived from biomass and soil-carbon inventories of Pan et al.<sup>[5]</sup>, thus providing additional support for a significant soil to hydrosphere anthropogenic carbon leakage. This budget analysis clearly identifies the need for an integrated process-based quantitative understanding of the LOAC response to anthropogenic perturbations, which include factors as diverse as land-use and climate change, hydraulic management, and agricultural, domestic and industrial activities. The anthropogenic perturbations affect both, the soil to hydrosphere C leakage and the fate of terrestrial C within the LOAC. Hydraulic management, for instance, is capable of changing the amount of C stored in sediments and to change the net-exchange of  $CO_2$  with the atmosphere. An integrated quantitative understanding of the LOAC response to the anthropogenic perturbations should rely on spatially-resolved contemporary budgets of lateral C transport, retention in sediments and loss through decomposition and degassing. Key environmental drivers of these fluxes need to be identified to assess their sensitivity with respect to global change and modeling efforts need to be pursued to support improved estimates of the anthropogenic perturbations are discussed in section 4 and 5 of this contribution.

#### 4. High resolution maps of CO<sub>2</sub> evasion from rivers

Observations of fluvial fluxes of solutes are a useful integrative measure for Earth Surface processes within a river catchment. Measurements of fluvial carbon fluxes, in particular, allow reassessing the C balance of terrestrial ecosystems by including the lateral carbon fluxes. A monitoring at the outlet of a river basin is, however, not sufficient because organic C and CO<sub>2</sub> stemming from soil respiration are not conservative during lateral transport and net-fluxes of CO<sub>2</sub> from the rivers to the atmosphere are significant<sup>[15]</sup>. At regional and global scales, a spatially explicit assessment of CO<sub>2</sub> evasion from the river is thus required to improve the contemporary terrestrial carbon budget. In a fully data driven approach, a dense network of water chemistry observations, from which CO<sub>2</sub> partial pressures (pCO<sub>2</sub>) and vertical river-atmosphere CO<sub>2</sub> fluxes (FCO<sub>2</sub>) could be calculated, would be necessary. Such monitoring network is currently not available as only industrialized countries have sufficient data coverage. Large parts of the globe remain only sparsely or not at all covered, including hotspot areas such as tropical Africa and Asia

and Siberia<sup>[11]</sup>.

In an ongoing study (Lauerwald et al. in prep), an empirical model for river pCO<sub>2</sub> was developed to produce a global river pCO<sub>2</sub> map at half a degree resolution. Compared to Raymond et al.,  $2013^{[15]}$ , which identifies broad regional patterns in global CO<sub>2</sub> evasion, this research thus takes the next step forward. Combining the pCO<sub>2</sub> map with spatially explicit estimates on stream surface area (A<sub>river</sub>) and gas exchange velocity (k) relying on empirical equations from the literature<sup>[15,16]</sup>, the first global FCO<sub>2</sub> map was created, at a resolution compatible with that of Earth System Models. The empirical pCO<sub>2</sub> model was trained on 1182 sampling locations with suitable time-series, covering all latitudes and biomes from the high north down to the central Tropics. The identified predictors of pCO<sub>2</sub> are average net primary production (NPP, g C m<sup>-2</sup>yr<sup>-1</sup>), population density (PD, inhabitants per km<sup>2</sup>), slope gradient within the river catchment (S, in °), and average air temperature at the sampling location (T, in °C).

In contrast to the predictor S, which is negatively correlated to river  $pCO_2$  and will not change significantly over the next centuries, the other three predictors have a positive effect on river  $pCO_2$  and are predicted to change considerably during the 21<sup>st</sup> century. Projections of these drivers for the year 2100 can thus be used to investigate plausible alterations of the global river  $CO_2$  evasion. Expected increases in PD and T, and partly NPP due to  $CO_2$ fertilization, will likely promote an increase in river  $pCO_2$ . The projected increase in atmospheric  $pCO_2$  FCO<sub>2</sub> could partly alleviate this effect by reducing the river-atmosphere  $pCO_2$  gradient and FCO<sub>2</sub>. Changes in river discharge resulting from modifications in precipitation patterns will entrain changes in A<sub>river</sub>, which will also have a direct effect of FCO<sub>2</sub>.

The predicted FCO<sub>2</sub> map produced for present-day conditions is used here to revisit the C budget of the Amazon River, as an example. The FCO<sub>2</sub> from streams and rivers is estimated at  $95 \pm 25$  TgC yr<sup>-1</sup> (Lauerwald et al., in prep). In a recent study, Abril et al.<sup>[17]</sup> estimated that floodplains of the central Amazon Basin contribute to a FCO<sub>2</sub> of  $210 \pm 60$  TgC yr<sup>-1</sup>. Together with the 29 TgC yr<sup>-1</sup> of DOC and 18 TgC yr<sup>-1</sup> POC exported to the Atlantic Ocean [Global NEWS2 <sup>[18]</sup>], the total export of C originally fixed by plants adds up to about 350 TgC yr<sup>-1</sup>, which represents about 8.5 % of the terrestrial NPP within the Amazon basin. However, a significant proportion of the FCO<sub>2</sub> from the floodplains but also from adjacent rivers was reported to originate from the root respiration of semi-aquatic, emergent vegetation on the floodplains<sup>[17]</sup>, and the contribution of terrestrial NPP to FCO<sub>2</sub> is thus likely lower. Projections on NPP, T, PD, atmospheric pCO<sub>2</sub> and river discharge will be used to provide a first-order estimate of how FCO<sub>2</sub> in the Amazon Basin might change until 2100.



Fig 1: Modelled average river pCO<sub>2</sub>, stream surface area (A<sub>river</sub>), and CO<sub>2</sub> evasion from rivers (FCO<sub>2</sub>) in the Amazon River Basin for the period 1990-2010. Units of FCO<sub>2</sub> refer to total continental areas, incl. inland waters.

#### 5. Perspectives: Including fluvial C transport into the land surface scheme ORCHIDEE

Fluvial C transport is currently ignored in Earth System models (ESMs) and the terrestrial C balance thus only considers NPP, soil heterotrophic respiration and land carbon storage. The integration of C leakage in ESMs will improve the allocation of the land and ocean C sinks and will allow attributing changes in lateral C fluxes to environmental drivers. In addition, the modelled fluvial C exports could also be used as input for global ocean biogeochemistry models. This objective, complementary to the empirical approach described above, requires an update of the land surface scheme ORCHIDEE by integrating the fluvial transfer of dissolved organic C (DOC) and dissolved  $CO_2$  from soils to the ocean, including DOC decomposition and  $CO_2$  evasion to the atmosphere during lateral transport (Fig.2).



Fig. 2: Schematic representation of the modelled water fluxes in ORCHIDEE. Geometrically, the river routing scheme (a) is organized in grid boxes (ig), which are further subdivided into basins (ib). It attributes, for each basin, the influxes of water and dissolved C received from neighboring basins. For each basin, the different reservoirs store and exchange water are defined (b). Blue arrows represent water fluxes from or to the reservoirs. ET: evapotranspiration, E: evaporation, I: infiltration, P: precipitation.

ORCHIDEE was recently upgraded to better represent soil organic C (SOC) pools and SOC decomposition (Guenet and Camino-Serrano, pers.com.). In particular, the soil hydrology module was updated to include DOC and CO<sub>2</sub> export from soil drainage (fig. 2). The river routing scheme of ORCHIDEE has now been enabled for DOC and  $CO_2$  transport with the water flow. For each basin, the routing scheme distinguishes three main dynamic reservoirs for water, DOC and dissolved CO<sub>2</sub>: the 'stream reservoir' (the river itself), the 'fast reservoir' (surface runoff) and the 'slow reservoir' (soil drainage). The routing scheme also represents the water exchange between the stream and the floodplain reservoirs, which are important for river systems like the Amazon<sup>[17]</sup>. At each time step, a fraction of the DOC stored in the reservoirs is converted to CO2 based on a reservoir-specific decomposition rate. The CO2 evasion from the rivers and floodplains is then calculated. The ORCHIDEE enabled for lateral C transport is currently calibrated for the Amazon, a basin for which the hydrology has already been calibrated and validated<sup>[19]</sup>. Ultimately, ORCHIDEE enabled for lateral C transport could be used to estimate the cumulative global land carbon storage change since the beginning of the Industrial Era. In the 5<sup>th</sup> assessment report of the IPCC<sup>[4]</sup>, a value of  $30 \pm$ 45 PgC was reported for this accumulation. Although it is a robust measurement-based estimate calculated as the residual of the other terms (anthropogenic emissions, atmospheric CO<sub>2</sub> growth rate and measured ocean C content change), the cumulative land C storage could actually be significantly smaller because of the continuous displacement of anthropogenic carbon through the terrestrial-aquatic interface.

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