Anthropogenic perturbation of the carbon fluxes from land to ocean

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A substantial amount of the atmospheric carbon taken up on land through photosynthesis and chemical weathering is transported laterally along the aquatic continuum from upland terrestrial ecosystems to the ocean. So far, global carbon budget estimates have implicitly assumed that the transformation and lateral transport of carbon along this aquatic continuum has remained unchanged since pre-industrial times. A synthesis of published work reveals the magnitude of present-day lateral carbon fluxes from land to ocean, and the extent to which human activities have altered these fluxes. We show that anthropogenic perturbation may have increased the flux of carbon to inland waters by as much as 1.0 Pg C yr\(^{-1}\) since pre-industrial times, mainly owing to enhanced carbon export from soils. Most of this additional carbon input to upstream rivers is either emitted back to the atmosphere as carbon dioxide (\(-0.4\) Pg C yr\(^{-1}\)) or sequestered in sediments (\(-0.5\) Pg C yr\(^{-1}\)) along the continuum of freshwater bodies, estuaries and coastal waters, leaving only a perturbation carbon input of \(-0.1\) Pg C yr\(^{-1}\) to the open ocean. According to our analysis, terrestrial ecosystems store \(-0.9\) Pg C yr\(^{-1}\) at present, which is in agreement with results from forest inventories but significantly differs from the figure of \(-1.5\) Pg C yr\(^{-1}\) previously estimated when ignoring changes in lateral carbon fluxes. We suggest that carbon fluxes along the land–ocean aquatic continuum need to be included in global carbon dioxide budgets.

During the past two centuries, human activities have greatly modified the exchange of carbon and nutrients between the land, atmosphere, freshwater bodies, coastal zones and the open ocean\(^1\). Together, land-use changes, soil erosion, liming, fertilizer and pesticide application, sewage-water production, damming of water courses, water withdrawal and human-induced climatic change have modified the delivery of these elements through the aquatic continuum that connects soil water to the open ocean through rivers, streams, lakes, reservoirs, estuaries and coastal zones, with major impacts on global biogeochemical cycles.\(^2\)–\(^14\) Carbon is transferred through the aquatic continuum laterally across ecosystems and regional geographic boundaries, and exchanged vertically with the atmosphere, often as greenhouse gases (Box 1).

Although the importance of the aquatic continuum from land to ocean in terms of its impact on lateral C fluxes has been known for more than two decades\(^1\),\(^5\),\(^6\), the magnitude of its anthropogenic perturbation has only recently become apparent.\(^8\),\(^11\),\(^12\),\(^16\),\(^18\). The lateral transport of C from land to sea has long been regarded as a natural loop in the global C cycle unaffected by anthropogenic perturbations. Thus, this flux is at present neglected in assessments of the budget of anthropogenic CO\(_2\) reported, for instance, by the Intergovernmental Panel on Climate Change (IPCC) or the Global Carbon Project.\(^19\)–\(^23\) Quantifying lateral C fluxes between land and ocean and their implications for CO\(_2\) exchange with the atmosphere is important to further our understanding of the mechanisms driving the natural C cycle along the aquatic continuum\(^24\),\(^25\), as well as for closing the C budget of the ongoing anthropogenic perturbation.

Data related to the C cycle in the aquatic continuum from land to ocean are too sparse to provide a global coverage, with insufficient water sampling, poorly constrained hydrology and surface area extent of various ecosystems, and few direct pCO\(_2\) and other carbon-relevant measurements.\(^26\),\(^27\). Global box models have been used to explore the magnitude of these fluxes and their anthropogenic perturbations, but the processes remain highly parameterized.\(^7\) The current generation of three-dimensional Earth system models includes the coupling between the C cycle and the physical climate system, but ignores lateral flows of C (and nutrients) altogether.\(^28\) Major challenges in the study of C in the aquatic continuum include the disentangling of the anthropogenic perturbations from the natural transfers, identifying the drivers responsible for the ongoing changes and, ultimately, forecasting their future evolution, for example, by incorporating these processes in Earth system models. Resolving these issues is not only necessary to refine the allocation of greenhouse-gas fluxes at the global and regional scale, but also to establish policy-relevant regional budgets and mitigation strategies.\(^29\)

The term ‘boundless carbon cycle’ was introduced to designate the present-day lateral and vertical C fluxes to and from inland waters only.\(^17\). Here, we extend this concept to all components of the global C cycle that are connected by the land–ocean aquatic continuum (Box 1) and discuss possible changes relative to the natural C cycle by providing new separate estimates for the present day and the anthropogenic perturbation. This distinction is important because, in some instances, bulk fluxes have been compared with perturbation fluxes, such as the net land C sink of anthropogenic CO\(_2\), which may cause confusion.\(^17\),\(^30\). Here we deal with the total C fluxes, but do not systematically distinguish between inorganic and organic, as this is still poorly known at the global scale for several of the components of the land–ocean continuum. However, we do highlight the exact chemical composition where it is sufficiently well constrained. Supplementary Table S1 is a compilation of the major flux estimates from the literature and estimated in this paper with a measure of confidence involving transfer of C from one global domain to another. A brief justification of our proposed estimate for each of these fluxes is also provided.

Contemporary estimates of lateral carbon fluxes

In this section, we derive contemporary estimates of the carbon fluxes along the continuum of the spectrum of land–ocean aquatic systems. We first look at the C transports involving inland waters and then consider their links to C flows through estuaries and the coastal ocean and beyond.

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Box 1 | Land–ocean carbon flux concepts

The land–ocean aquatic continuum. This can be represented as a succession of chemically and physically active biogeochemical systems, all connected through the continuous water film that starts in upland soils and ends in the open ocean. Carbon is transferred along this continuum. These systems are often referred to as filters, because carbon is not only transferred, but also processed biogeochemically and sequestered in sediments or exchanged with the overlying atmosphere as greenhouse gases (Fig. 1a).

The pre-industrial land–ocean loops. Lateral carbon transfer through the aquatic continuum was already active under pre-industrial conditions and the boundless carbon cycle consists of two loops. The organic carbon loop starts with the lateral leakage of some of the organic carbon that is fixed into the terrestrial biosphere by photosynthesis. This carbon is then transferred horizontally through aquatic channels down to the coastal and open ocean where C is returned to the atmosphere as CO$_2$. The inorganic loop is driven by the land-based weathering of silicate and carbonate rocks that consumes atmospheric CO$_2$, and the subsequent transport of the weathering products of cations, anions and dissolved inorganic carbon to the ocean, where part of the CO$_2$ is returned to the atmosphere through ocean carbonate sediment formation (a process that increases pCO$_2$ in seawater). The other part is returned by volcanism. Both loops are generally assumed to have been in a quasi-steady-state initial condition in pre-industrial times, that is, they were globally balanced at the millenial timescale.

Anthropogenic perturbation of the lateral carbon fluxes. Human perturbations to the lateral carbon fluxes have moved the boundless carbon cycle away from this global balance, causing imbalances in the fluxes and stocks, such as the C inputs from soils to inland water systems, the strength of the terrestrial ecosystem C cycle (Box 1) and represents about 5% of soil C based on upscaling of local C budgets$^{17,26}$. This input is composed of four fluxes. The first and largest one is soil-derived C that is released from soil respiration ($F_s$), which releases an additional $\sim$0.1 Pg C yr$^{-1}$ as an input to freshwaters$^{39,41}$. The fourth flux involves photosynthetic C fixation within inland waters, potentially high on an areal basis$^{46}$. A substantial fraction of this C is returned to the atmosphere owing to decomposition within inland waters$^{32}$, but a percentage remains for export and burial$^{43,44}$, and priming of terrestrial organic matter decomposition$^{46}$. Thus, although aquatic systems can emit CO$_2$ to the atmosphere, they still can be autotrophic$^{46}$. We estimate with low confidence that 20% of the organic C buried and exported from inland waters (see below for estimations) is autochthonous ($F_A$).

Physical erosion of particulate inorganic C ($\sim0.2$ Pg C yr$^{-1}$) and of organic C that is resistant to mineralization ($\sim0.1$ Pg C yr$^{-1}$) represents another C source to the aquatic continuum$^{17,28}$ ($F_E$). Although the fate of this physically eroded C is difficult to trace, it is likely to be refractory at the centennial timescale$^{48}$ and is most likely channelled through inland waters and estuaries to the open ocean without significant exchange with the atmosphere. It is thus treated separately in Fig. 1a.

During the transport of C from soils to the coastal ocean, a fraction of the lateral flux that transits through inland waters is emitted to the atmosphere, mainly as CO$_2$ ($F_E$). CH$_4$ is also emitted from lakes and some rivers ($F_l$), but this flux represents a small fraction of the laterally transported C flux$^{30}$. Data-driven estimates of the water-to-atmosphere CO$_2$ efflux have been obtained for individual components of the inland freshwater continuum$^{16,17,50}$. This efflux is sustained by CO$_2$ originating from root and soil respiration, aquatic decomposition of dissolved and particulate organic matter, and decomposition of organic C from sewage, as detailed above. Furthermore, the addition of C from fringing and riparian wetlands, counted as soil C input to freshwaters in Fig. 1a, may also contribute significantly to freshwater CO$_2$ outgassing$^{51}$. Approximately 12,000 sampling locations of the inorganic C cycle are now reported in inland water databases (Fig. 1a). Calculation of CO$_2$ from alkalinity and pH indicates that 96% of inland waters are oversaturated with respect to CO$_2$, relative to its atmospheric concentration, while 82% have a concentration of at least twice that of the atmosphere (Global River Chemistry Database (GloRiCh), unpublished data; ref. 52).

Numerous measurements of the freshwater CO$_2$ efflux are available for some regions of the globe, such as the Rhine catchment, Scandinavia and the conterminous United States$^{39,42,51–53}$. However, lack of direct CO$_2$ flux measurements, incomplete spatial coverage of CO$_2$ sampling locations coupled with the difficulty in determining the surface area of inland waters, and scaling the gas-transfer velocity in freshwaters, causes large uncertainties and prevents us from obtaining robust global-scale estimates (Fig. 2a). In particular, many rivers and lakes that contribute a significant fraction to the aquatic C flux remain poorly surveyed in terms of pCO$_2$ (GloRiCh, unpublished data). These include the rivers of Southeast Asia, tropical Africa and the Ganges and, to a lesser extent, the waters of the Amazon Basin$^{34,55}$, which carry disproportionately high organic

Inland waters. The present-day bulk C input (natural plus anthropogenic) to freshwaters was recently estimated at 2.7–2.9 Pg C yr$^{-1}$, based on upscaling of local C budgets$^{17,26}$. This input is composed of four fluxes. The first and largest one is soil-derived C that is released from soil respiration ($F_s$) and net C fixation ($F_I$). The soil-derived C flux is part of the terrestrial ecosystem C cycle (Box 1) and represents about 5% of soil C based on upscaling of local C budgets$^{17,26}$. This input is composed of four fluxes. The first and largest one is soil-derived C that is released from soil respiration ($F_s$), which releases an additional $\sim$0.1 Pg C yr$^{-1}$ as an input to freshwaters$^{39,41}$. The fourth flux involves photosynthetic C fixation within inland waters, potentially high on an areal basis$^{46}$. A substantial fraction of this C is returned to the atmosphere owing to decomposition within inland waters$^{32}$, but a percentage remains for export and burial$^{43,44}$, and priming of terrestrial organic matter decomposition$^{46}$. Thus, although aquatic systems can emit CO$_2$ to the atmosphere, they still can be autotrophic$^{46}$. We estimate with low confidence that 20% of the organic C buried and exported from inland waters (see below for estimations) is autochthonous ($F_A$).
Figure 1 | Global carbon budget and its anthropogenic perturbation. Estimates are shown for (a) the present day (2000–2010), (b) the natural C cycle (>1750) and (c) the anthropogenic perturbation only. All fluxes are in Pg C yr⁻¹, rounded to ±0.05 Pg C yr⁻¹, and refer to total C fluxes (organic and inorganic C). The numbers associated with the arrows are fluxes between reservoirs. Boxed \( \Delta C \) refers to C accumulation within each reservoir. The red box delineates the succession of lateral C filters along the land–ocean aquatic continuum (LOAC). The + sign indicates that C sequestration from estuaries and adjacent coastal vegetation are merged in the figure. The stars in panel (a) indicate the confidence interval associated to the flux estimates, based on The First State of the Carbon Cycle Report. A black star means 95% certainty that the actual estimate is within 50% of the estimate reported; a grey star means 95% certainty that the actual value is within 100% of the estimate reported; a white star corresponds to an uncertainty greater than 100%. Flux symbols are defined in Table 1.
carbon loads owing to their combination of high terrestrial productivity, high decomposition rates and high uniform precipitation rates (Fig. 2a). The scarcity of direct pCO$_2$ measurements and lack of knowledge on regional surface area and gas-transfer velocity explain the large uncertainty in the CO$_2$ outgassing from freshwaters$^{16,26,31}$, with a range of 0.6–1.25 Pg C yr$^{-1}$. The values at the higher end of the spectrum also include the contribution of streams and small lakes, which are typically not considered in flux estimates$^{26}$. We estimate a most likely value of the CO$_2$ outgassing flux of 1.1 Pg C yr$^{-1}$ ($F_2$) with a medium-to-low confidence.

The burial rate in freshwater sediments has been estimated to be between ~0.2 and 1.6 Pg C yr$^{-1}$. The lower estimate refers to lakes, ponds and reservoirs only$^{16,26}$(0.2–0.6 Pg C yr$^{-1}$), whereas the upper one also includes sedimentation in floodplains$^{36,57}$(0.5–1.6 Pg C yr$^{-1}$). The factor of eight between the lower and higher bound estimates of this burial flux highlights the limited observational data available to constrain this term at the global scale. Within this large uncertainty, we adopt with a low confidence a value of 0.6 Pg C yr$^{-1}$ for the C burial in inland water sediments ($F_3$). Part of this burial is carbon transported, by erosion processes, from soils to lake sediments and floodplains.

From the mass balance of the C input from soils to fresh waters minus CO$_2$ outgassing and C burial fluxes in inland waters adopted here, the output represents a lateral C flux transported downstream into estuarine systems ($F_4$) of 1.0 Pg C yr$^{-1}$. Thus our estimate is close to values based on compilation of field data$^{47,58}$ and the results of the Global Nutrient Export from Watersheds model of carbon and water flows$^{26}$, although higher values have also been suggested$^{4}$. A flux of particulate and dissolved organic C, each equivalent to about 0.2 Pg C yr$^{-1}$, and a flux of dissolved inorganic C of about 0.4 Pg C yr$^{-1}$ is the conventional partitioning among the different C pools$^{43,54,64}$. If we take into account the uncertainty for each of the individual inland water fluxes (weathering, outgassing, burial and export), the balance also indicates that the soil-derived C flux ($F_5$, 1.9 Pg C yr$^{-1}$) is certainly not known any better than within ±1.0 Pg C yr$^{-1}$.

Estuaries. In our analysis, estuaries (total area of 1.1 x 10$^5$ km$^2$) correspond to the boundary between inland aquatic systems and the coastal ocean, represented mainly by the shelves of the world’s oceans$^{26}$. Recent syntheses of observational data indicate that estuaries emit CO$_2$ to the atmosphere$^{24,61}$, within the range of 0.25 ± 0.25 Pg C yr$^{-1}$ ($F_6$). Field measurements suggest that about 10% of the CO$_2$ outgassing from estuaries is sustained by the input from upstream freshwaters ($F_7$) and 90% by local heterotrophy$^{4}$, with a significant fraction of the required organic C coming from adjacent marsh ecosystems ($F_8$). Although coastal vegetated environments (salt marshes, mangroves, seagrasses, macroalgae and coral reefs) may export as much as 0.77–3.18 Pg C yr$^{-1}$ to the coastal ocean$^{62}$, we use here a more conservative estimate of 0.3 ± 0.1 Pg C yr$^{-1}$ for the common estuarine vegetation of mangroves and salt marshes, which is based on an upsampling of a detailed regional budget for the southeastern United States$^{63}$. Furthermore, to our knowledge, no global estimates exist for C burial in all estuarine sediments, but a long-term burial in mangroves and salt marshes of 0.1 ± 0.05 Pg C yr$^{-1}$ has been proposed$^{25,64}$ ($F_9$). If we combine our upstream river and coastal vegetation inputs with our average estuarine CO$_2$ outgassing estimate to the atmosphere and the first-order estimate for burial of C in estuarine sediments and vegetated ecosystems ($F_8$), we obtain a C delivery from estuaries to the coastal ocean of 0.95 Pg C yr$^{-1}$ ($F_{10}$). This estimate amounts to about one-third of the initial C flux released from soils, rocks and sewage as input to inland freshwater systems.

Coastal ocean and beyond. Materials leaving estuaries transit into the coastal ocean and beyond to the open ocean. Recent syntheses of the air–sea CO$_2$ fluxes in coastal waters (total area of 31 x 10$^5$ km$^2$)$^{26}$ suggest that at present between 0.22 and 0.45 Pg C yr$^{-1}$ are taken up by the coastal ocean$^{25,64}$. We choose here a lower estimate of 0.2 Pg C yr$^{-1}$ for the coastal ocean sink of CO$_2$, based on a recent analysis for the global ocean$^{69}$ ($F_1$). This value relies on the observation that, outside the nearshore environments, the net CO$_2$ fluxes in the coastal regions are of similar strengths and directions to those in the adjacent ocean regions, that is, that low-latitude coastal regions tend to be sources of CO$_2$ to the atmosphere, whereas high-latitude regions tend to be sinks$^{66,68}$. This allows the extrapolation of the open-ocean CO$_2$ exchange values towards the coasts. Although this extrapolation is an oversimplification, the most recent estimate (0.25 Pg C yr$^{-1}$) based on the upsampling from a few sites with good observational coverage suggests a similar value$^{68}$. Nevertheless, it is important to recognize that the limited spatial coverage of pCO$_2$ data in the coastal ocean (Fig. 2b) and its heterogeneous nature confine the confidence to low-to-medium. Furthermore, the influence of terrestrial C input on air–sea CO$_2$ fluxes extends considerably beyond the limit of the shelf in the discharge plumes of large tropical rivers, such as the Amazon$^{63,70}$. These plumes should be considered as an integral part of the land–ocean continuum.

Coastal ocean sediments may sequester between 0.2 and 0.5 Pg C yr$^{-1}$ of organic C and calcium carbonate$^{1,72}$, although significantly higher values have been reported$^{72}$ ($F_{11}$). We choose here a central estimate of 0.35 Pg C yr$^{-1}$, of which a sediment C burial of

| Symbol | Flux name |
|--------|-----------|
| FF     | Fossil-fuel emissions |
| FT$_1$ | Net primary production of terrestrial ecosystems |
| FT$_2$ | Harvest, cattle and biofuels emissions (CO$_2$) |
| FT$_3$ | Fire emissions (CO$_2$) |
| FT$_4$ | Cattle, landfills and fire emissions (CH$_4$) |
| FT$_5$ | Terrestrial biomass to soil C flux |
| FT$_6$ | Rice paddies and wetland emissions (CH$_4$) |
| FT$_7$ | Soil heterotrophic respiration (CO$_2$) |
| F$_1$  | Total soil C input to inland waters |
| F$_2$  | Inorganic C input to inland waters from weathering |
| F$_3$  | Atmospheric C uptake by bedrock weathering |
| F$_4$  | Organic C inputs to inland waters (sewage) |
| F$_5$  | Photosynthetically fixed C not respired in inland waters |
| F$_n$  | Physical erosion of total recalcitrant C |
| F$_6$  | CH$_4$ emissions from inland waters to the atmosphere |
| F$_7$  | CO$_2$ emissions from inland waters to the atmosphere |
| F$_8$  | Total C burial in inland water sediments |
| F$_9$  | Total lateral C flux from inland waters to estuaries |
| F$_{10}$ | CO$_2$ emissions from estuaries to the atmosphere |
| F$_{11}$ | CO$_2$ uptake from marsh ecosystems and subsequent organic C input to estuaries |
| F$_{12}$ | Total C burial in estuarine sediments and coastal vegetated ecosystems |
| F$_{13}$ | Total lateral C flux from estuaries to the coastal ocean |
| F$_{14}$ | Atmospheric C uptake by coastal waters |
| F$_{15}$ | Total C burial in coastal ocean sediments |
| F$_{16}$ | Total C export from the coastal to the open ocean |
| FO$_1$ | Air-sea CO$_2$ flux in the open ocean |
| FO$_2$ | Total C burial in open ocean sediments |
| FG     | Geologic fluxes (volcanism, metamorphism and oxidation of fossil organic C) |
0.05–0.1 Pg C yr\(^{-1}\) is attributed solely to the seagrass meadows of shallow coastal seas\(^{18}\). In addition, the most probable repository for much of the recalcitrant terrestrial C related to physical weathering (\(F_R\)) is likely to be in coastal sediment C pools\(^{74,75}\). Furthermore, the net pumping of anthropogenic CO\(_2\) from the atmosphere into coastal waters may increase the dissolved inorganic carbon storage in the water column\(^{76}\), by about 0.05 Pg C yr\(^{-1}\). Because of data paucity, a direct global estimate of lateral C fluxes at the boundary between the coastal and open ocean, delineated by the shelf break\(^{62}\), is not at present achievable solely through observational means\(^{12,70,77}\). Thus, based on mass-balance calculations using the above flux estimates, we propose with a low confidence that the net inorganic and organic C export from the coastal ocean to the open ocean is \(\sim 0.75\) Pg C yr\(^{-1}\) (\(F_w\)) as shown in Fig. 1a.

**Anthropogenic perturbation of lateral carbon fluxes**

As with the contemporary lateral C fluxes, we now trace sequentially the route of the perturbed C fluxes through the global system of inland waters to estuaries to coastal waters and beyond.

**Inland waters.** Reconstructions of the historical evolution (pre-industrial, around the year 1750, to present) of the global aquatic C cycle and its fluxes have so far relied primarily on globally averaged box models\(^2,24\). Increasing concentrations of atmospheric CO\(_2\), land-use changes, nitrogen and phosphorus fertilizer application, C, nitrogen and phosphorus sewage discharge and global surface temperature change drive these highly parameterized models. Model simulations suggest that the transport of riverine C (\(F_R\)) has increased by about 20% since 1750, from \(\sim 0.75\) Pg C yr\(^{-1}\) in 1750 to 0.9–0.95 Pg C yr\(^{-1}\) at present. The existence of such an enhanced riverine delivery of C is supported by the available literature data\(^{3,8,47,74}\), and has been attributed to deforestation and more intensive cultivation practices that have increased soil degradation and erosion. This leads to an increase in the export of organic and inorganic C

![Figure 2](https://www.biogeomod.net/geomap)
to aquatic systems. For example, erosion of particulate organic C in the range 0.4–1.2 Pg C yr\(^{-1}\) has been reported for agricultural land alone.\(^{15,16}\) However, only a percentage of this flux represents a lateral transfer of anthropogenic CO\(_2\) fixed by photosynthesis.\(^{16,69-70}\)

Although budgets have been established for present-day conditions,\(^{16,20,21}\) there is no observationally based estimate of the pre-industrial C flux from soils to inland waters, nor of the associated CO\(_2\) outgassing and C burial fluxes in freshwater systems in pre-industrial times. Furthermore, we are not aware of any spatially explicit model simulation of the CO\(_2\) outgassing and C burial fluxes in inland aquatic systems during the industrial period at the global scale. The potential anthropogenic effects on C cycling in various inland aquatic systems have been reviewed,\(^{16}\) but a quantitative estimate of the anthropogenic perturbation remains to be assessed. The bulk fluxes are nevertheless large enough that even a small change would alter the global C budget of anthropogenic CO\(_2\).

For example, it is highly likely that damming and freshwater withdrawal have impacted the CO\(_2\) outgassing fluxes and organic carbon burial rates since pre-industrial time through their effect on surface area and residence time of inland waters.\(^{2,8,9}\) In particular, the evolution in agricultural practices and the construction of human-made impoundments during the past century have most likely led to enhanced CO\(_2\) outgassing. A CO\(_2\) evasion of 0.3 Pg C yr\(^{-1}\) for human-made reservoirs alone has been reported.\(^{82,83}\) Furthermore, a C burial flux in the sediments of reservoirs and small agricultural ponds of 0.35 Pg C yr\(^{-1}\) has also been estimated\(^{84-89,91-93}\) with C probably from terrestrial and autochthonous sources.

To estimate the extent to which other inland water environments such as lakes, streams and rivers have been perturbed by human activities, we assume that CO\(_2\) outgassing and C burial fluxes in these systems linearly scale with the estimated increase (~20%) in soil-derived C exported from rivers to estuaries (\(F_2\)) and the coastal zone (see also above). This leads to a perturbation of ~0.1 Pg C yr\(^{-1}\) for the CO\(_2\) outgassing flux and ~0.05 Pg C yr\(^{-1}\) for the C burial flux. The linear scaling assumption implies that CO\(_2\) outgassing and the C sedimentation rate are first order processes with respect to the additional C concentration derived from enhanced exports from soil in the freshwater aquatic systems. This assumption is probably reasonable for the air–water flux, but the change in C burial flux is almost surely more complex.\(^{41}\)

Sewage inputs to upstream rivers (\(F_4\)) are inferred to add another 0.1 Pg C yr\(^{-1}\) to the anthropogenic perturbation, and we make the assumption that this labile organic C is entirely outgassed within inland waters. Combining all contributions, the budget analysis gives CO\(_2\) outgassing (\(F_1\)) and C burial (\(F_5\)) fluxes under pre-industrial conditions of 0.6 and 0.2 Pg C yr\(^{-1}\), respectively (Fig. 1b).

The remaining extra outgassing (0.5 Pg C yr\(^{-1}\)) and extra burial (0.4 Pg C yr\(^{-1}\)) fluxes are then attributed to the anthropogenic perturbation (Fig. 1c). Furthermore, increased chemical weathering of continental surfaces caused by human-induced climate change and increased CO\(_2\) levels contributes to the enhanced riverine export flux of C derived from rock weathering (\(F_3\)). The anthropogenic perturbation could possibly reach 0.1 Pg C yr\(^{-1}\), mainly through enhanced dissolution of carbonate rocks.\(^{85}\) The impact of land-use change on weathering rates may have started 3,000 years ago but its effect on atmospheric CO\(_2\) is difficult to assess.\(^{85,86}\) For example, C mobilized from the practice of agricultural liming is a source of enhanced land-use C fluxes\(^{85}\) and could result in a sink of ~0.05 Pg C yr\(^{-1}\).

Summing up, the total present-day flux from soils, bedrock and sewage to aquatic systems of 2.5 Pg C yr\(^{-1}\) shown in Fig. 1a can be decomposed as the sum of a natural flux of ~1.5 Pg C yr\(^{-1}\) (Fig. 1b) and an anthropogenic perturbation flux of ~1.0 Pg C yr\(^{-1}\) (Fig. 1c) — a value that is similar to a previously published estimate. Roughly 50% of this anthropogenic perturbation (0.5 Pg C yr\(^{-1}\)) is respired back to the atmosphere in freshwater systems (\(F_6\)), while the remainder contributes to enhanced C burial (\(F_8\)) and export to estuaries (\(F_9\)) and, perhaps, to the coastal ocean (\(F_{13}\), Fig. 1c).

**Estuaries.** The perturbation of historical drainage and human-assisted conversion of salt marshes and mangroves, as well as the channelization of estuarine conduits, have modified the estuarine C balance. For instance, the total loss of C from these intertidal C pools could be as high as 25–50% over the past century, mainly because of land-use change.\(^{44}\) Assuming that the reduction in the C flux from marshes and mangrove ecosystems to estuaries (\(F_{11}\)) is proportional to the reduction in the surface area of these ecosystems, we estimate that the pre-industrial flux of C transported from coastal vegetation to estuaries must have been about 0.15 Pg C yr\(^{-1}\) larger than that of the present-day value of 0.30 Pg C yr\(^{-1}\). We predict that C burial in estuarine sediments has been reduced from pre-industrial times to the present by the same relative factor, amounting to an anthropogenic reduction of 0.05 Pg C yr\(^{-1}\) of the estuarine sediment C burial flux (\(F_{12}\)) in Fig. 1b. In the absence of independent evidence, we assume that the air–sea estuarine flux of CO\(_2\) has remained constant since pre-industrial times (\(F_{10}\), Fig. 1c). With the constraints mentioned above, closing the mass balance of the pre-industrial and present-day estuarine C budgets requires that the C export to the coastal ocean (\(F_{13}\)) has increased by ~0.1 Pg C yr\(^{-1}\) since 1750, from 0.85 to 0.95 Pg C yr\(^{-1}\).

**Coastal ocean and beyond.** Lacking sufficient observational evidence, we have to rely on process-based arguments and models to separate present-day C fluxes for the coastal ocean into pre-industrial and anthropogenic components. Perhaps the best constrained flux component is the uptake of anthropogenic CO\(_2\) across the air–sea interface, which has been estimated to about 0.2 Pg C yr\(^{-1}\), on the basis that this uptake has the same flux density as that of the mean ocean, namely about 6 g C m\(^{-2}\) yr\(^{-1}\). This assumption is warranted as the oceanic uptake flux of anthropogenic CO\(_2\) is to first order controlled by the surface area. Much less certain is the degree to which the enhanced nutrient and C inputs to the coastal ocean could have modified the air–sea CO\(_2\) balance. Box model simulations for the global coastal ocean suggested that the enhanced supply of nutrients from land may have increased coastal productivity and C burial in coastal sediments,\(^{40}\) from about 0.2 Pg C yr\(^{-1}\) to 0.35 Pg C yr\(^{-1}\), as well as contributing to a substantial increase in the air-to-sea CO\(_2\) flux,\(^{12,14}\) by up to 0.2–0.4 Pg C yr\(^{-1}\). However, the efficiency by which the additional nutrient supply delivered to the coastal ocean is actually reducing the CO\(_2\) budget is globally uncertain. For example, on continental shelves, the enhanced supply of nitrogen (<50 Tg N yr\(^{-1}\))\(^{94-95}\) may stimulate a maximal additional growth of about 0.3 Pg C yr\(^{-1}\), of which only a portion is exported to depth, and of which less than 50% is released by uptake of CO\(_2\) from the atmosphere.\(^{96}\) We estimate that coastal eutrophication has caused an increase in the air–sea CO\(_2\) flux no larger than ~0.1 Pg C yr\(^{-1}\). The response of the highly heterogeneous, very shallow coastal ocean, including reefs, banks and bays (<50 m, 12 x 10\(^{6}\) km\(^{2}\))\(^{94}\), remains largely unknown. However, it is in this region that the nutrient impact on biological productivity, organic C burial and area-specific CO\(_2\) fluxes is expected to be the highest. Therefore, the anthropogenic air–coastal C burial flux is only known with low confidence. We estimate a conservative value of 0.2 Pg C yr\(^{-1}\) for this anthropogenic flux (\(F_{14}\)), which is significantly lower than the value of 0.5 Pg C yr\(^{-1}\) suggested in recent syntheses.\(^{70}\)

The fate of the additional C received from the estuaries (\(F_{13}\)) is unclear. Some of this C is probably sequestered in coastal sediments, together with some of the additional organic C produced in response to the nutrient input, amounting to a flux potentially as large as 0.1–0.15 Pg C yr\(^{-1}\) (\(F_{14}\)) The remainder is exported to the open ocean, together with some of the anthropogenic CO\(_2\), taken up from the atmosphere, amounting to a flux of approximately
heterotrophy of many systems, but cannot yet be quantified by enhanced decomposition of autochthonous organic materials exported to the open ocean and the remaining >30% is emitted to estuarine and coastal sediments, <20% is fixed in inland waters, at present approximately 50% is sequestered in inland water, estuarine and coastal sediments, <20% is emitted to the atmosphere–terrestrial ecosystems CO₂ sink, (2) burial of displaced carbon downstream into freshwater and coastal sediments, (3) outgassing of CO₂ to the atmosphere en route between land and the coastal ocean, and (4) a net open ocean carbon storage increase larger than the atmospheric CO₂ flux. For clarity, reservoirs to atmospheric fluxes of CO₂ are in black, lateral fluxes of carbon displaced at the surface are in green, and changes in (anthropogenic) carbon storage are given in red. In the figure, TES_{BCC} = RLS_{SCE} + FEO_{BCC} – COU_{BCC}. The‘net atmosphere–terrestrial ecosystems CO₂ sink’ is equal to TES_{BCC} = LUC. The ‘net anthropogenic CO₂ outgassing’ for the freshwater–estuary–coastal zone continuum is FEO_{BCC} – COU_{BCC}. Uncertainties on fluxes are from the GCP, except for the LOAC fluxes (identified by an asterisk), where indicative estimates are given based on the categorization proposed in The First State of the Carbon Cycle Report and for TES_{BCC} (see below). The same applies for the uncertainty on the LOAC carbon storage change. The uncertainty on TES is calculated from the atmospheric mass balance, assuming quadratic errors propagation, and a similar approach is used for the uncertainty on carbon storage (ΔC) for the terrestrial ecosystems and open ocean, based on their respective mass balance. The uncertainty associated to each LOAC flux was estimated using the categories proposed in The First State of the Carbon Cycle Report. These categories have then been converted to uncertainty values assuming that they follow a Gaussian error distribution. σ = μ/4: 95% certain that the estimate is within 50% of the proposed value. σ = μ/2: 95% certain that the estimate is within 100% of the proposed value. σ > μ/2: uncertainty greater than 100%. μ = mean value.

0.1 Pg C yr⁻¹ (F_{co2}). This value is again significantly lower than previous estimates⁹, highlighting that our confidence in these numbers is very low.

In summary, although accurate quantification remains challenging, one can firmly conclude that during the industrial era, the laterally transported C fluxes and the vertically exchanged atmospheric CO₂ fluxes relevant to the land–ocean aquatic continuum have been significantly altered by human activities, the main driver being land-use changes. Our analysis suggests that out of the ~1.1 Pg C yr⁻¹ of extra anthropogenic C delivered to the continental to land–ocean aquatic systems (0.8 Pg C yr⁻¹ from soils, 0.1 Pg C yr⁻¹ from weathering, 0.1 Pg C yr⁻¹ from sewage, 0.1 Pg C yr⁻¹ from enhanced C fixation in inland waters), at present approximately 50% is sequestered in inland water, estuarine and coastal sediments, <20% is exported to the open ocean and the remaining >30% is emitted to the atmosphere as CO₂. CO₂ fluxes along the land–ocean continuum may not only be altered directly by increased anthropogenic C export from soil and subsequent respiration, but also indirectly by enhanced decomposition of autochthonous organic materials triggered by priming. This indirect process may be a quantitatively relevant contribution to the estimated fluxes and the observed net heterotrophy of many systems, but cannot yet be quantified⁶⁹¹. The uncertainties associated with our breakdown are large and represent a fundamental obstacle for global C cycle assessments and a fertile avenue for future research (see also Fig. 3). Future work that succeeds in narrowing down the uncertainties on the anthropogenic perturbations may overrule our conclusions on the qualitative value of each flux in the analysis, but is unlikely to affect our conclusion that the anthropogenic perturbation to the aquatic continuum C fluxes is important for the global carbon budget.

Implications for the global carbon budget
Quantitative assessment of the C fluxes through the land–ocean aquatic continuum in a so-called boundless C cycle analysis has important implications for how terrestrial C fluxes ought to be evaluated and how the sinks of anthropogenic CO₂ over land and ocean need to be attributed. This assessment has implications for terrestrial ecosystem C cycling, global terrestrial and ocean C models, atmospheric inversions, ocean C inventories and the global anthropogenic CO₂ budget. The land C cycle (see Supplementary Note for further details) is driven by the C input to ecosystems due to net primary productivity of ~59 Pg C yr⁻¹ (FTₚ, Fig. 1a). A small fraction of net primary productivity is used by ecosystems to increase C stocks, as evidenced by the net growth of many forests⁶². Humans and fires consume another fraction (FTₙ and FTₕ), and litter production (FTₗ) is thus actually lower now than what it was for the natural
cycle. After some time of residency in ecosystems, most of the C is returned to the atmosphere as CO$_2$ by heterotrophic respiration (FT$_r$), while a small fraction is channelled to freshwaters.

In the majority of global terrestrial ecosystem model formulations, the lateral C fluxes from soils to freshwaters are not represented, and modellers assume one-dimensional closure of C between terrestrial ecosystem pools and the atmosphere. Consequently, soil heterotrophic respiration is overestimated in these models relative to observations. Similarly, global ocean biogeochemistry models use prescribed lateral input of C (or nutrients) from land as an input for the open ocean. At present, their coarse resolution does not resolve the coastal ocean well, and their simple, globally tuned formulations of ecosystem and biogeochemical processes may not be able to capture fully the complexity of the impacts of the enhanced terrestrial inputs of C and nutrients on the coastal ocean.

Atmospheric CO$_2$ inversion models estimate regional scale net land–atmosphere CO$_2$ fluxes from CO$_2$ concentration gradients measured by surface network stations. Thus, the lateral transport of C at the surface is not a process generally considered in inversion modelling, which only detects vertical CO$_2$ fluxes. Inversion estimates of land–atmosphere CO$_2$ fluxes do include CO$_2$ exchange with inland waters and estuaries in their regional output. However, the spatial resolution of inversion CO$_2$ exchange estimates is too coarse, and the atmospheric sampling too sparse to separate CO$_2$ fluxes from inland waters from those exchanged by terrestrial ecosystems. The same caveat applies for atmospheric inversions of the air–sea CO$_2$ fluxes. These inversion approaches evaluate the net flux across the air–sea interface, which includes the effect of the lateral and vertical C exchanges along the aquatic continuum, in particular the net outgassing of the riverine carbon.

Changes in the open-ocean C inventory over the historical period have been used to infer the cumulated oceanic C sink. Most recently, a global oceanic storage of anthropogenic C of 155 ± 30 Pg C for the period from 1800 to 2010 has been estimated$^{44}$. This storage includes only that part of the C that has been taken up through the air–sea interface in response to the increase in atmospheric CO$_2$, that is, the anthropogenic CO$_2$. Not included in this oceanic net C sink estimate is any additional air–sea CO$_2$ flux that was driven by other anthropogenically driven processes, such as coastal nutrient inputs and consequent enhanced productivity and burial of organic carbon. If we take our estimate of ~0.1 Pg C yr$^{-1}$, and assume that it can be scaled in time with the increase in atmospheric CO$_2$ concentrations, this might have caused an additional oceanic C storage of 10 Pg C over the industrial era that needs to be added to the global increase in oceanic storage.

In the global CO$_2$ budget reported for instance by the IPCC and by the Global Carbon Project (Fig. 3a) the ‘land residual’ sink is deduced as a difference between fossil-fuel and land-use-change emissions of C and atmospheric accumulation and open-ocean uptake$^{21,22,95}$, the ocean component being estimated from forward and inverse models$^{21,95}$. This method implicitly assumes that the pre-anthropogenic global carbon budget was at steady state and the fluxes along the land–ocean aquatic continuum, unlike most other fluxes, have no anthropogenic component. Thus, these ‘classical’ budgets ignore the anthropogenic perturbation of the boundless carbon cycle displayed in Fig. 1c. Our new estimation of these fluxes allows us to deconvolute the ‘land residual sink’ into (1) a ‘terrestrial ecosystem sink’ of anthropogenic CO$_2$ comprising the contribution of the land vegetation, litter, soils and the bedrock, and (2) sources and sinks of anthropogenic CO$_2$ occurring in the aquatic ecosystems of the freshwater–estuarine–coastal ocean continuum (Fig. 3b). We find that the ‘terrestrial ecosystem sink’ in the boundless carbon cycle is removing ~2.85 Pg C yr$^{-1}$ of anthropogenic CO$_2$ from the atmosphere (Fig. 3b). This sink of CO$_2$ is larger than the residual land sink estimates reported by the IPCC or the Global Carbon Project$^{21}$ (Fig. 3a) because a fraction of this flux is returned to the atmosphere by CO$_2$ outgassing along the ecosystems of the land–ocean aquatic continuum. However, only 0.9 Pg C yr$^{-1}$ of this terrestrial ecosystem sink is actually sequestered in biomass and soil of land ecosystems, as 1.0 Pg C yr$^{-1}$ is released to the atmosphere owing to land-use changes, and a similar amount (1.0 Pg C yr$^{-1}$) is exported to the land–ocean aquatic continuum. The net biomass and soil sequestration estimate calculated here is consistent with the ‘bottom-up’ estimates reported in Fig. 1c from biomass and soil-carbon inventories$^{95}$ (0.8 Pg C yr$^{-1}$), thus providing additional support to our independent estimation of the anthropogenic C delivered to the water continuum (see Supplementary Note). Enhanced rock weathering contributes also to the anthropogenic perturbation (Fig. 3b, 0.1 Pg C yr$^{-1}$). The anthropogenic C delivered to freshwaters is partly outgassed to the atmosphere as CO$_2$ (0.55 Pg C yr$^{-1}$), partly sequestered in sediments (0.35 Pg C yr$^{-1}$) and partly exported to the coastal ocean (0.1 Pg C yr$^{-1}$). The coastal ocean also contributes to the anthropogenic CO$_2$ budget (0.2 Pg C yr$^{-1}$ air–sea uptake, 0.2 Pg C yr$^{-1}$ sequestered in coastal sediments and water column).

Critical quantifications

Although we have demonstrated that it is possible to establish closed C and anthropogenic CO$_2$ budgets, broadly consistent with the current growth rate of atmospheric CO$_2$, the component fluxes for the land–ocean aquatic continuum cannot be adequately quantified through a robust statistical treatment of available datasets yet. The data are also too sparse to resolve fully the diversity of soil types, inland waters, estuaries and coastal systems. In particular, wetland and riparian ecosystems lateral fluxes are not known. Nevertheless and importantly, revised anthropogenic CO$_2$ budgets need to consider and assess quantitatively the anthropogenic perturbation to the aquatic continuum. Any improved estimates will certainly at least require a considerably denser carbon and CO$_2$ flux observation system, based on direct measurements of CO$_2$ gas-transfer velocities and ecosystem surface areas. These measurements should be completed by an expansion of pCO$_2$ sampling and, in some cases, of flux towers into wetlands and aquatic systems, to have continuous monitoring. Areas of regional priority include the Amazon and the Congo riverine basins and their tropical coastal currents. The Ganges River system, the Bay of Bengal, the Indonesian Archipelago, the Southeast Asian

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seas and the Arctic rivers are other critical regions having significantly large carbon inputs into the coastal seas. Furthermore, a quantitatively mechanistic understanding of the key processes controlling CO$_2$ outgassing and preservation of C in the land–ocean continuum is needed. An important unknown involves knowledge of the sources, transport pathways and liability and rates of degradation of accumulating organic and inorganic C, be it in soils, the aquatic system or the sea floor. The mechanistic understanding is necessary to parameterize the various processes involving C and their sensitivity to external perturbations at the larger scales of Earth system models. At present, this lack of understanding limits our ability to predict the present and future contribution of the aquatic continuum fluxes to the global C budget involving anthropogenic CO$_2$.

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Additional information

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Additional information

Competing financial interests

The authors declare no competing financial interests.
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