A Review on the Riverine Carbon Sources, Fluxes and Perturbations

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Abstract Carbon is transported from the land to the oceans via rivers and groundwater. The transfer of organic matter from the land to the oceans via fluvial systems is a key link in the global carbon cycle. Rivers also provide a key link in the geological scale carbon cycle. Nevertheless, an appreciation of their roles is yet to be made. Even when their roles are included, data are drawn only from selected large rivers, often neglecting the small mountainous rivers. Previous studies have demonstrated that, the tropic rivers, especially located in Asian region play crucial role in regulating the global carbon budgets. Superimposed on the natural sources and fluxes, the anthropogenically-induced fluxes, primarily emanating from reduced sediment and discharge (as a result of constructions of dams and reservoirs), and enhanced detrital organic matter (as a result of increased surface flow due to land use change) introduce perturbations.

Keywords Carbon cycle · River transport · Sources and fluxes of carbon · Perturbations

1 Introduction

Global biogeochemical cycles have shaped the Earth's climate and surface environment since the earliest days of the planet. Carbon in the biosphere is unevenly distributed among three major reservoirs: terrestrial, oceanic and atmospheric. Simplified depictions of the global carbon cycle have generally consisted of two biologically active boxes (oceans and land) connected through gas exchanges with a third box, the atmosphere (Bolin 1981; Siegenthaler and Sarmiento 1993; IPCC 2007). As models developed further, more sub-compartments and processes have been added in an attempt to unravel the more intricate interactions among them

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(for example, Parton et al. 1994; Foley et al. 1996; Canadell et al. 2000; Cramer et al. 2001). Work during the 1970s and 1980s demonstrated that rivers deliver significant amounts of terrestrially-derived organic and inorganic C from land to the sea (Degens et al. 1991; Schlesinger and Melack 1981). This riverine "pipe" transports C from land to the ocean. When inland aquatic systems are included in global models, it is usually only for the transport of C through the riverine pipe. This delivery of terrestrial C through the riverine drainage network is in fact the end result of a number of transformations and losses in aquatic systems en route.

In the global carbon cycle, rivers have a critical role in connecting terrestrial, oceanic and atmospheric carbon reservoirs. Atmospheric Carbon is transported by rivers from terrestrial ecosystems as soil dissolved and particulate organic C (Dissolved Organic Carbon or DOC, Particulate Organic Carbon or POC) and Dissolved Inorganic C (DIC) and supplied to the oceans. Terrestrial organic matter thus represents approximately one third of the organic matter buried in all marine sediments and is stored over geological timescales leading to atmospheric carbon dioxide sequestration (Berner 1989). Of the portion of riverine that originates from the atmosphere, organic carbon is formed by the photosynthesis reaction; the fraction of atmospheric carbon, that is, dissolved inorganic carbon (DIC) comes from soil CO_2, fixed from the atmosphere by the weathering of rocks and air-water exchange. Of the terrestrial portion, DIC and particulate inorganic carbon (PIC) are associated with the weathering of rock.

Given cognizance to the importance of the fluvial system and the dynamics within drainage basins on the global carbon cycle, this paper reviews the importance of rivers in collecting, sequestering, transporting and delivering carbon to other realms of the Earth system.

2 Sources and Transport of Riverine Carbon

Carbon is transported from the land to the oceans via rivers and groundwater. The transfer of organic matter from the land to the oceans via fluvial systems is a key link in the global carbon cycle. Rivers also provide a key link in the geological scale carbon cycle by moving weathering products to the ocean. But organic carbon does not passively through the river systems; even very old and presumably recalcitrant soil carbon may be partially remineralized in aquatic systems. Remineralization of organic carbon during transport leads to elevated levels of dissolved CO_2 in rivers, lakes and estuaries worldwide. These high concentrations subsequently lead to outgassing to the atmosphere on the order of 1 Pg C year^{-1} with the majority in the humid tropics.

The most obvious source of export of C from the continental margins occurs through riverine flux. These fluxes are large, fairly well quantified, and have been derived from estimates of water discharge (for example, Dai and Trenberth 2002) and measurements of aqueous carbon concentrations. River export of organic carbon to the sea has thus been estimated as ranging from 0.38 (Degens et al. 1991) to 0.53 Pg C year^{-1} (Stallard 1998) with several other estimates falling within this range (for example, Schlesinger and Melack 1981; Meybeck 1982; Ludwig et al. 1996, b; Aitkenhead and McDowell 2000). Riverine export of dissolved inorganic carbon resulting from the fixation of atmospheric carbon through rock weathering is likely to be between 0.21 and 0.3 Pg C year^{-1} (Stallard 1998). Globally about half of the bicarbonate transported by rivers originates from silicate weathering (in which case 100 % of the bicarbonate came from CO_2 sequestration) and half from carbonate weathering (in which case only half the bicarbonate came from CO_2 sequestration (Stallard 1998; Meybeck 1993).

Groundwater export to the sea has not been considered as yet in global C budgets. Groundwater comprises 97 % of the world's liquid freshwater (van der Leeden et al. 1990) and can contain substantial quantities of organic and inorganic carbon (Cai 2003; Hem 1985). Some groundwater discharges as the base flow of rivers and is included in river carbon export. However, estimates of submarine groundwater discharge (SGD) span a broad range (Church 1996; Cai 2003). Imbalances in the world water budget (van der Leeden et al. 1990; Dai and Trenberth 2002; Shiklomanov and Rodda 2003) and groundwater residence times from 3 to 25 ka suggest SGD equal to 1.4–12 % of river influx, with the most accepted values between 5 and 10 % (Taniguchi et al. 2002; Slomp and Van Cappellen 2004). Estimates of groundwater alkalinity of around 60 mg^{-1} (Cai 2003) and a minimum DOC concentration of 1 mg^{-1} (Simpkins and Parkin 1993) suggest SGD of carbon of 0.13–0.25 Pg C year^{-1}. Collectively, using mid-range values for the river and groundwater components, inland waters thus deliver about 0.9 Pg C year^{-1} to the oceans, roughly equally as inorganic and organic carbon.

3 Types of Fluxes

Large rivers tend to integrate the biogeochemical activities within the drainage basin and the total carbon observed in river water is a mixed component that originates from different sources. In a pristine environment, the basic nature of riverine carbon consists of three categories: (a) Dissolved inorganic carbon (DIC) derived from chemical weathering of rocks which is largely transported as HCO_3^- ion, (b) Particulate organic carbon (POC) derived from soil organics, litter fall and autochthonous production; and (c) Dissolved organic carbon (DOC) arising from leaching of top-soil, peat and regulated by in situ pH. (Sarin et al. 2002).

This input from rivers is composed of four fluxes. The first and largest one is soil-derived C that is released to inland waters, mainly in organic form (particulate and dissolved), but also as free dissolved CO_2 from soil respiration (Sarmiento and Sundquis 1992). The flux is estimated to be 1.9 Pg C year^{-1}, by subtracting, from a total median estimate of 2.8 Pg C year^{-1}, the smaller contributions from the other three fluxes: chemical weathering, sewage and net C fixation. The soil-derived C flux is part of the terrestrial ecosystem C cycle and represents about 5 % of soil heterotrophic respiration. Current soil respiration estimates neglect the C released to inland waters. A downward revision of the estimate of soil heterotrophic respiration to account for the soil C channeled to inland freshwater systems would nevertheless remain within the uncertainty of this flux (Meybeck 1982). The second flux involves the chemical weathering of continental surfaces (carbonate and silicate rocks). It is part of the inorganic (often called ‘geological’) C cycle and causes an additional ~0.5 Pg C year^{-1} input to upstream rivers (Beusen et al. 2005; Schlesinger and Melack 1981; Borges and Abril 2012; Laruelle et al. 2010). About two-thirds of this C flux is due to removal of atmospheric CO_2 in weathering reactions and the remaining fraction originates from chemical weathering of C contained in rocks. The pathway for chemical weathering is nevertheless largely indirect with most of the CO_2 removed from the atmosphere being soil CO_2, having passed through photosynthetic fixation. Weathering releases C to the aquatic continuum in the form of dissolved inorganic C, mainly bicarbonate, given that the average pH is in the range of 6–8 for freshwater aquatic systems (Chen and Borges 2009).

In contrast to soil-derived organic C, it is assumed that C derived from rock weathering will not degas to the atmosphere during its transfer through inland waters (Borges et al. 2005). Over geological timescales, silicate weathering coupled with carbonate precipitation in the Ocean is responsible for a large fraction of atmospheric CO_2 sequestration that balances the mantle and metamorphic CO_2 inputs into the atmosphere and therefore regulates the global climate (e.g. Walker et al. 1981; Berner et al. 1983). While modern weathering rates are often derived from river solute fluxes (e.g. Meybeck 1987; Gaillardet et al. 1999a; West et al. 2005) their solid counterparts have received far less attention (e.g. Gaillardet et al. 1999b; France-Lanord and Derry 1997; Gislason et al. 2006) probably because of the difficulty of integrating the variability of detrital sediments over space and time (Lupker et al. 2011; Bouchez et al. 2010, 2011a, b). Sediment records are however one of the rare archives that can be reliably used to trace past erosion fluxes at regional scales.

The third flux represents the C dissolved in sewage water originating from biomass consumption by humans and domestic animals, which releases an additional ~0.1 Pg C year^{-1} as an input to freshwaters (Borges 2005). The fourth flux involves photosynthetic C fixation within inland waters, potentially high on an areal basis. A substantial fraction of this C is returned to the atmosphere owing to decomposition within inland waters (Duarte et al. 2005) but a percentage remains for export and burial (Breithaupt et al. 2012) and priming of terrestrial organic matter decomposition (Liu et al. 2010). Thus, although aquatic systems can emit CO_2 to the atmosphere, they still can be autotrophic.

4 Role of Chemical Weathering in Regulating the Carbon Cycle

Chemical weathering of silicate rocks consumes significant quantities of CO_2 that has regulated the global carbon cycle and in so doing Earth’s climate over several eons (Arvidson et al. 2006; Berner 2004; Kempe and Degens 1985; Walker et al. 1981).

The carbon dioxide in the atmosphere dissolves in rainwater forming carbonic acid, which, once in contact with rocks, slowly dissolves them. This atmospheric carbon is then transported by rivers into the oceans, where it is trapped for several 1,000 years, before returning to the atmosphere or alternatively being stored in marine sediments or in organisms secreting aragonite/calcite shells and tests. During the weathering of silicate rocks, the totality of the HCO_3^- ions released in solution comes from atmospheric/soil CO_2 as it can be noted for example in the albite hydrolysis:

$$2NaAlSi_3O8 + 2CO_2 + 11H_2O \; --- > A_{l2}SiO_5(OH)_4 + 2HCO_3^- + Na^+ + 4H_4SiO_4$$

Considering the weathering of carbonate rocks, only half of the HCO_3^- ions released in solution come from the atmospheric/soil CO_2 as it can be seen in the calcite dissolution:

$$CaCO_3 + CO_2 + H_2O \rightarrow Ca^{++} + 2HCO_3^-$$

Chemical weathering is central in surface biogeochemical cycles because it redistributes the chemical elements between Earth's surface reservoirs such as continental crust and the Ocean. Over geological timescales, silicate weathering coupled with carbonate precipitation in the Ocean is responsible for a large fraction of atmospheric CO_2 sequestration that balances the mantle and metamorphic CO_2 inputs into the atmosphere and therefore regulates the global climate (e.g. Walker et al. 1981; Berner et al. 1983). Continental weathering consumes about 0.3 Gt of atmospheric CO_2 each year (Gaillardet et al. 1999a, b).

Chemical weathering of rock helps regulate the supply of nutrients and solutes to soils, streams and the ocean, and is also the long-term sink for atmospheric CO_2, thus modulating Earth's climatic evolution via the greenhouse effect. Thus, to the extent that chemical weathering rates increase with temperature, weathering feedbacks should, over millions of years, buffer Earth's climate against large temperature shifts (Riebe et al. 2004). This chemical weathering process stores around 0.3 billion tons of atmospheric carbon in rivers and in the oceans every year: although this is considerably less than human-induced CO_2 production (around 8 billion tons per year), it is roughly equivalent to the net exchange flux between the atmosphere and the terrestrial biosphere (vegetation, soil, humus, etc.) under preindustrial conditions (0.4 billion tons). The long-term cooling effect is due to the higher weatherability of basalts compared to granite. In the same climatic conditions, basaltic surfaces consumes between 5 and 10 times more atmospheric CO_2 than granitic surfaces. Despite this, chemical weathering of the continents has never been taken into account until now in models of future climate change.

5 Current Understanding on the Riverine Carbon Transport

Transport of material via rivers is under study for over 100 years in geochemical budgets. It gives essential information both on processes affecting the continental surface (weathering, plant production, pollution etc.) and on the amount and nature of material carried to water bodies such as lakes, seas and oceans. Many world-wide budgets have been published based on ever increasing studies of river dissolved and particulate material (Martin and Meybeck 1979; Meybeck 1979). Two approaches are available for estimating global fluvial carbon fluxes. One uses carbon data for large rivers in various regions. For instance, Meybeck (1979) estimated global DIC and PIC fluxes (0.38 and 0.17 Pg C/year) based on data for 60 large rivers or groups of rivers that together are responsible for 63 % of global river discharge and considered runoff and average watershed temperature to obtain information regarding the other 37 %. Ludwig et al. (1996) utilized a database of mean annual dissolved organic carbon (DOC) and particulate organic carbon (POC) fluxes of 29 and 19 rivers respectively and other ecological factors to calculate DOC and POC fluxes (0.21 and 0.17 Pg C/year, respectively). The main determinants of DOC fluxes are the drainage intensity, basin slope and amount of organic soil carbon. The main factors that govern POC fluxes are the total mass of suspended matter (TSM) and sediment load. The other approach considers the mass balance. For example, Mackenzie et al. (1998) evaluated fluvial inorganic and organic carbon fluxes (0.72 and 0.61 Pg C/year) using a conceptual model. Notably, published results considered only the total quantity of inorganic or organic carbon (in the case of Mackenzie et al. 1998) or only some of the four carbon components (in the case of Meybeck 1979 and Ludwig et al. 1996). Importantly, the IPCC (2007) report considered only DIC and DOC fluxes. As reported by various studies worldwide, the amount of terrigenous carbon that enters the river systems is on the order of 1.5 Pg C a^{-1} (range: 0.8–2 Pg C a^{-1}). The present-day bulk C input (natural plus anthropogenic) to freshwaters was recently estimated at 2.7–2.9 Pg C $year^{-1}$, based on upscaling of local C budgets (Raymond et al. 2008).

Studies generally focus on large river systems like the Mississippi, the Ganga–Brahmaputra, Amazon or large Arctic rivers which integrate differences in lithology, vegetation, soils and climate. Small mountainous rivers directly connected to the oceans are less studied than large rivers, although they play an important role in transport of organic matter, their yields and runoff being inversely proportional to the watershed area (Milliman and Meade 1983). Recent works have demonstrated that these small rivers are major sources of POC, DOC and dissolved major elements to the oceans. Due to their location in the tropical zone, numerous small mountainous rivers are affected by periodic intense precipitation events such as cyclones or tropical storms that can play an important role on soil erosion and can potentially increase total organic carbon fluxes released by these systems. Gaps exist in the understanding of spatial and temporal variations of organic carbon and their fluxes from the tropical rivers, which are less studied because of their location in the developing countries. This is in spite of their high water discharge (>60 %) and 34 % of total suspended load supply into the global oceans (Martin and Meybeck 1979; Meybeck 1988; Ludwig et al. 1996; Ludwig and Probst 1998; Balakrishna and Probst 2005).

Particulate inorganic carbon concentrations in tropical rivers are negligible because of the dissolution of PIC into DIC during the weathering of carbonate rocks (Sarin et al. 2002). Tropical rivers provide 0.53 Pg C/year of riverine carbon to the oceans, of which 39.8 % is DIC, 25.7 % is DOC, 9.7 % is PIC and 24.8 % is POC. The largest DIC flux within the tropical region is found in Asia, because the DIC concentration is highest there and the discharge is second highest. The Americas have the highest DOC flux in the tropical area, owing to the sheer volume of discharge. The highest PIC flux in tropical regions is also found in the Americas because they have the highest PIC/TSM ratio and the second highest sediment load. Asia has the highest specific carbon yields in the tropical region, because of the high ratios of discharge to surface area, and sediment load to surface area. Anthropogenic activities, however, such as reducing sediment load and increasing the amount of detrital organic matter in rivers, may continue to change the fluvial carbon fluxes of tropical rivers. In India scattered studies have been made on the organic carbon. For example, Sarin et al. (2002) studied the DOC, POC and DIC concentrations on Godavari, the largest peninsular Indian tropical river.

6 Anthropogenic Perturbation of the Carbon Fluxes

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 (Likens et al. 1981; Mulholland. and Elwood 1982; Wollast and Mackenzie 1989; Degens et al. 1991; Smith and Hollibaugh 1993; Stallard 1998; Ver et al. 1999; Richey 2004; Raymond et al. 2008). 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 (Mackenzie et al. 2005; Cotrim da Cunha et al. 2007; Quinton et al. 2010). Carbon is transferred through the aquatic continuum laterally across ecosystems and regional geographic boundaries and exchanged vertically with the atmosphere, often as greenhouse gases.

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, the magnitude of its anthropogenic perturbation has only recently become apparent. 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. 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, as well as for closing the C budget of the ongoing anthropogenic perturbation. Superimposed on these natural forms, the present-day increased amounts of industrial effluents, fertilizers, sewage and other human wastes are modulating the riverine concentrations of carbon. Anthropogenic changes and river eutrophication is an important factor in future for algal POC, which can create near-anoxic conditions when reaching coastal waters.

Land use change is currently extremely rapid and its consequences are more evident in the tropical regions, in part because of the disproportionate share of human population growth that is taking place in the tropics. Land clearing and conversion causes substantial loss of carbon and nitrogen and a lesser loss of sulphur and phosphorus from cleared sites in most regions. Climate change and increased nutrient deposition from the atmosphere will affect soils, plant productivity and biogeochemical cycles. The overall emphasis of the biogeochemistry is the terrestrial regulation of element pools, transformation gains and losses as they are altered by components of global change. In addition, there are a number of regions in which land-use and atmospheric composition and anticipated climate change are likely to alter the biogeochemistry of terrestrial ecosystems significantly and consequently to cause significant change on the riverine biogeochemistry.

Model simulations suggest that the transport of riverine C has increased by about 20 % since 1750, from ~0.75 Pg C year^{-1} in 1750 to 0.9–0.95 Pg C year^{-1} at present. The existence of such an enhanced riverine delivery of C is supported by the available published data (Milliman and Meade 1983; Meybeck 1982; Wollast and Mackenzie 1989; Richey 2004; Richey et al. 1991) 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 to aquatic system (Raymond et al. 2008). For example, erosion of particulate organic C in the range 0.4–1.2 Pg C year^{-1} has been reported for agricultural land alone (Stallard 1998; Quinton et al. 2010). However, only a percentage of this flux represents a lateral transfer of anthropogenic CO_2 fixed by photosynthesis (Stallard 1998; Smith et al. 2001; Billings et al. 2010).

7 Conclusions

- The carbon cycle plays an important role in regulating interactions among the lithosphere, hydrosphere, atmosphere, and biosphere. An understanding on the transfer of C between these spheres is essential from the environmental point of view. Rivers act as conduits in transporting, sequestering, and delivering C from lithosphere to ocean basins.

- At present, there is substantial lacuna in understanding of the sources, transport pathways and rates of C transfer to different spheres which inhibits our ability to predict the present and future contribution of the riverine fluxes to the global C budget involving anthropogenic CO_2.
- The extremely complex nature of riverine systems, due to variations in climate, land use, soil composition, hydrology and man's impact, complicates interpretation of relative roles and contributions of geogenic and anthropogenic sources and fluxes of carbon. Therefore, adequate characterization of such complex systems requires measurement of a number of parameters over extended periods of time.

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