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Gaseous and fluvial carbon export from an Amazon forest watershed

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Abstract The transfer of carbon (C) from Amazon forests to aquatic ecosystems as CO_2 supersaturated in groundwater that outgases to the atmosphere after it reaches small streams has been postulated to be an important component of terrestrial ecosystem C budgets. We measured C losses as soil respiration and methane (CH_4) flux, direct CO_2 and CH_4 fluxes from the stream surface and fluvial export of dissolved inorganic C (DIC), dissolved organic C (DOC), and particulate C over an annual hydrologic cycle from a 1,319-ha forested Amazon perennial first-order headwater watershed at Tanguro Ranch in the southern Amazon state of Mato Grosso. Stream pCO_2 concentrations ranged from 6,491 to 14,976 μatm and directly-measured stream CO_2 outgassing flux was 5,994 ± 677 g C m^{-2} y^{-1} of stream surface. Stream pCH_4 concentrations ranged from 291 to 438 μatm and measured stream CH_4 outgassing flux was 987 ± 221 g C m^{-2} y^{-1}. Despite high flux rates from the stream surface, the small area of stream itself (970 m^2, or 0.007% of watershed area) led to small directly-measured annual fluxes of CO_2 (0.44 ± 0.05 g C m^2 y^{-1}) and CH_4 (0.07 ± 0.02 g C m^2 y^{-1}) per unit watershed land area. Measured fluvial export of DIC (0.78 ± 0.04 g C m^{-2} y^{-1}), DOC (0.16 ± 0.03 g C m^{-2} y^{-1}) and coarse plus fine particulate C (0.001 ± 0.001 g C m^{-2} y^{-1}) per unit watershed land area were also small. However, stream discharge accounted for only 12% of the modeled annual watershed water output because deep groundwater flows dominated total runoff from the watershed. When C in this bypassing groundwater was included, total watershed export was 10.83 g C m^{-2} y^{-1} as CO_2 outgassing, 11.29 g C m^{-2} y^{-1} as fluvial DIC and 0.64 g C m^{-2} y^{-1} as fluvial DOC. Outgassing fluxes were somewhat lower than the 40–50 g C m^{-2} y^{-1} reported from other Amazon watersheds and may result in part from lower annual rainfall at Tanguro. Total stream-associated gaseous C losses were two orders of magnitude less than soil respiration (696 ± 147 g C m^{-2} y^{-1}), but total losses of C transported by water comprised up to about 20% of the ± 150 g C m^{-2} (±1.5 Mg C ha^{-1}) that is exchanged annually across Amazon tropical forest canopies.

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Introduction

Streams and small rivers are important links between terrestrial and aquatic ecosystems because they receive inputs of carbon (C) as dissolved inorganic C (DIC), dissolved organic C (DOC) and particulate C (POC) from terrestrial ecosystems and then transform these materials at land-stream interfaces and in stream channels as water moves to larger rivers (McClain et al. 2003; Richey et al. 2009). Understanding the fate of these different forms of C in linked terrestrial-aquatic systems is important for understanding the controls on stream and river metabolism and the transport of C in rivers and C fluxes to oceans (Raymond and Bauer 2001; Caraco and Cole 2004). Small freshwater lakes and streams tend to be sources of CO_2 to the atmosphere because they receive inputs of groundwater that is supersaturated in CO_2 derived from adjoining terrestrial sources, and that CO_2 is subsequently released to the atmosphere as outgassing (Telmer and Vezier 1999; Jones et al. 2003; Hanson et al. 2004). Streams are also the location of respiration of terrestrially-derived dissolved and particulate C (Dillon and Molot 1997; Rantakari and Kortelainen 2005; Sobek et al. 2005).

The land-to-water transport and subsequent potential outgassing of C carried in groundwater has received increasing attention as an important component of watershed C budgets (Cole et al. 2007). Outgassing of CO_2 from streams has been estimated to account for up to 50% of net annual C accumulation in arctic tundra (Kling et al. 1991) and up to 70% of C accumulation in peatlands (Hope et al. 2001). In the Amazon, Richey et al. (2002) estimated that outgassing from rivers in the central floodplain region was roughly equal to estimated net annual C accumulation of 1.2 Mg C ha^{-1} y^{-1} in moist *terra firme* tropical forest. Extrapolated across the Amazon basin, this outgassing flux was approximately 10 times greater than fluvial export of organic C to the ocean in the Amazon River and roughly of similar magnitude to C loss or accumulation in the range of $\pm$ 1.5 Mg C ha^{-1} y^{-1} estimated for Amazon forest from recent eddy flux and biometric measurements (Meir et al. 1996; Phillips et al. 1998, 2009; Saleska et al. 2003; Malhi et al. 2004; Miller et al. 2004; Vourlitis et al. 2004).

Understanding the magnitude of aquatic outgassing and fluvial transport of C is important for understanding the magnitude and direction of the Amazon forest as a component of the global C budget. Evidence for potential outgassing of CO_2 comes from direct measurements of high pCO_2 concentrations in both large and small rivers of the Amazon. River waters of major tributaries of the Amazon basin are supersaturated in CO_2 with respect to the atmosphere, with values ranging to more than 12,000 ppmv and averaging 4,330 ppmv (Richey et al. 2002). The concentrations in small streams can be higher. Johnson et al. (2008) measured pCO_2 concentrations of up to 48,700 µatm and frequently higher than 25,000 µatm at the source of a small Amazon forest stream. Davidson et al. (in press) found pCO_2 frequently >10,000 µatm in three streams of mixed land use in the eastern Amazon. Johnson et al. (2008) also found that concentrations dropped to <5,000 ppmv within 20 m of the stream source, indicating rapid outgassing as groundwater comes in contact with the atmosphere. While these concentrations indicate the potential for transfer to the atmosphere of C produced by respiration in terrestrial soil, the total area of stream surfaces from which these transfers occurs is small.

Recent evidence from the Amazon suggests that riverine C fluxes as DOC and POC are small in comparison to outgassing, but these also have been measured directly in few locations and are poorly known. Johnson et al. (2006a) estimated that the transport of DIC from headwater streams on the Brazilian Shield in Mato Grosso was 40 times that of DOC and that DOC and POC export combined amounted to 4.9 g C m^{-2} y^{-1}. Waterloo et al. (2006) estimated slightly higher DOC export of 19.0 g C m^{-2} y^{-1} from a lowland stream in the central lowland Amazon.

In contrast to fluxes of CO_2, fluxes of CH_4 from streams and rivers are generally lower, but CH_4 fluxes can be important because methane is a powerful greenhouse gas with a global warming potential approximately 26 times that of CO_2 (Solomon et al. 2007). Tropical soils can serve as both sources and sinks of CH_4, depending primarily on soil moisture,

land use and other anthropogenic activities, but forest soils generally are net sinks for CH_4 on an annual basis (Keller and Reiners 1994; Steudler et al. 1996; Keller et al. 1990), although the development of anoxic conditions, primarily in riparian zones and on floodplains, can result in high rates of CH_4 generation (Agostinetto et al. 2002). Supersaturation of CH_4 in stream channels has also been found, although links between CH_4 production in streams and a terrestrial C source is not clear (Devol et al. 1988; Richey et al. 1988; Jones and Mulholland 1998a, b). The role of small Amazon streams in outgassing of CH_4 is unknown.

We quantified the form and magnitude of outgassing and fluvial dissolved inorganic and organic C export from a small Amazon forest watershed in the state of Mato Grosso, Brazil. We measured: (1) pCO_2 of streamwater and direct outgassing of CO_2 from the stream surface, (2) fluvial export of DOC, DIC and POC, and (3) CO_2 and CH_4 fluxes from soils. We used the results to evaluate the relative importance of watershed outgassing and fluvial C fluxes and to compare streamwater C losses to watershed-scale C transfers from soil to the atmosphere.

Methods

Study site

The study was conducted at Tanguro Ranch (12° 53′ S, 52° 21′ W) in the southern Brazilian Amazon state of Mato Grosso. Tanguro Ranch lies southeast of the Xingu Indigenous Reserve in the municipalities of Canarana and Querência at an elevation of approximately 350 m above sea level in the upper region of the Xingu River drainage between the Tanguro and Darro Rivers. Topography of the region consists of broad, gently sloping upland plateaus away from the rivers and gentle slopes to the waterways. It lies on a portion of the Brazilian Shield underlain by Precambrian Gneisses of the Xingu Complex. Soils consist of Oxisols, according to USDA Soil Taxonomy (Haplustox/Latossolo vermelho-amarelo distrófico, according to Brazilian Soil Taxonomy) on the plateaus. Soils of the plateaus have a water table depth at 12–15 m with no impeding layers. They have a bulk density of 1.06–1.24 g cm^3, clay content of 33–55% and carbon content of 8.6 kg m^{-2} in the top 1 m (Neu 2009). Soils of the plateau grade into soils on hillslopes that have similar bulk density (0.99–1.28 g cm^{-3}), clay content (37–58%) and carbon content 9.9 kg m^{-2} in the top 1 m (Neu 2009). Wetter, regularly-inundated Inceptisols (Haplusteps/Gleissolos) occur in smaller areas at the river margins (Projeto Radambrasil 1981). These soils have a similar bulk density (1.09 g cm^{-3}), slightly lower clay content (29–44%) and higher carbon content (11.6 kg m^{-2}) in the top 1 m (Neu 2009).

The forest vegetation of the Tanguro region is evergreen tropical forest (Ivanauskas et al. 2004), typical of the transition from Cerrado to the central Amazon rainforest. This forest has lower species diversity, smaller trees and lower canopy heights compared with many previously-studied Amazon forests (Ivanauskas et al. 2004). The climate at Tanguro is humid tropical (Köppens Aw). Average annual rainfall is 1,905 ± 271 mm (1987–2007) and strongly seasonal, with <5% of total annual precipitation falling between May and September. June, July and August are the driest months with less than 10 mm rain per month. Mean annual temperature is 25°C.

We examined a forest watershed of 1,319 ha drained by a perennial first-order stream. Three transects (plateau, hillslope and lowland) were collected during a hydrological year within the forest and one gauging station was established at the exit of this watershed reach (Fig. 1). The watershed is dominated by a plateau typical of the region. Near the stream, topography forms a distinct shoulder hillslope zone and a flatter but narrow lowland zone near the stream channel. Stream flows are dominated by groundwater moving within a deep water table with little temporal variation over the year. The watershed boundaries were delineated from Shuttle Mission Radar Topography data and validated with field measurements. We measured widths of the stream channel (at base flow) and the hillslope and lowland zones by hand at 20 points at regular intervals along an 800 m stream reach downstream from a small seep that defined the beginning of the perennial stream channel.

Rainfall and stream discharge

We estimated rainfall and streamflow from 15 Jan 2007 to 15 Jan 2008. Rainfall was estimated from

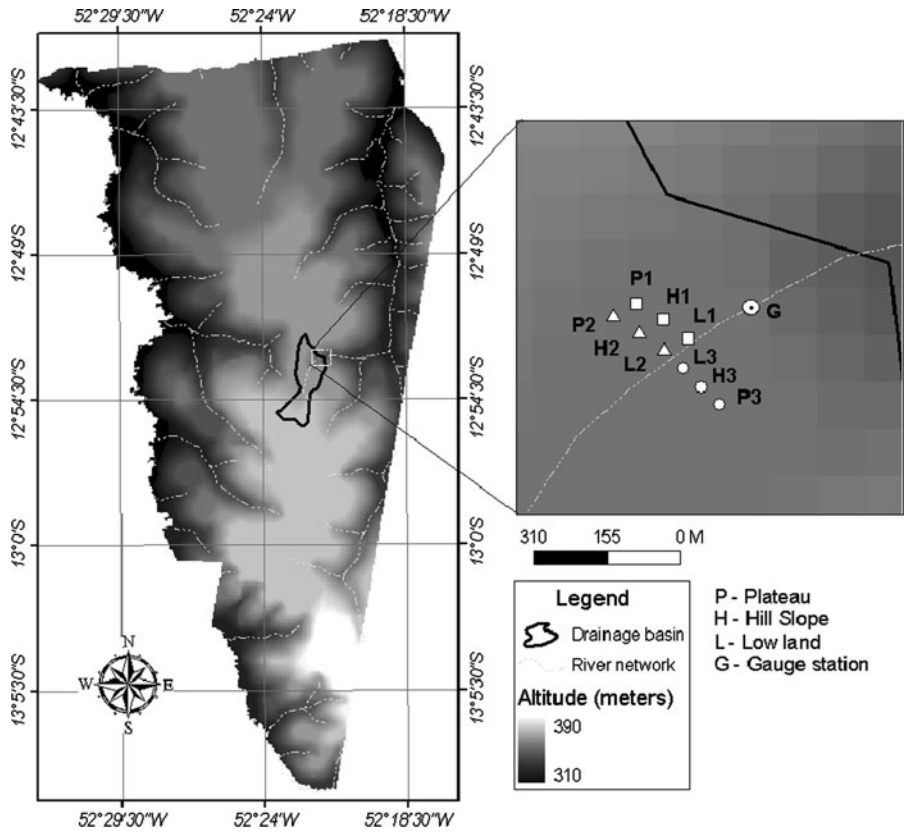

Fig. 1 Map of Tanguro Farm, showing the locations of the watershed and approximate sets of collectors in the three transects within the forest

several sources. Hourly rainfall and rain intensity were recorded at a weather station at the Tanguro Ranch field station 27 km south of the watershed sampling point. Daily rainfall was also recorded in two collectors 2 and 5 km from the watershed sampling point. Stream stage was recorded every 15 min from May to August 2007 with an ISCO 6200 automatic sampler equipped with a pressure transducer module. From August 2007 to August 2008 stream stage was also recorded hourly with an Onset water level. A rating curve ($y = 4.7986e^{-0.0483x}$, $r^2 = 0.95$) that related discharge to stream stage was developed by periodic measurements of stream discharge by measuring cross-sectional water velocities with a Global Water FP-101 current meter across a range of stream flows. We estimated hourly stream flows for January to May 2007 from daily rainfall and rainfall-streamflow relationships derived from the entire 16-month streamflow record beginning in May 2007. We estimated base and storm flows over the year using the local-minima hydrograph separation method (Sloto and Crouse 1996). This method separates peaks using a straight line between two local minima bounding precipitation maxima. Approximately once every 2 months during baseflow, discharge was also measured 800 m upstream of the primary sampling point and compared with logged discharge to estimate groundwater input in the reach.

Because flows of deep groundwater potentially bypass the stream gauging and sampling point, we used an energy balance model calibrated for the Tanguro region (Coe et al. 2009) to estimate annual evapotranspiration. We used that estimate, plus measured annual rainfall and streamflow to estimate the amount of water that left the watershed as deep groundwater flow that was not captured by our steam gauge.

Soil–atmosphere fluxes of CO_2 and CH_4

Measurements of soil-atmosphere flux were sampled from 14 February 2007 to 15 January 2008. We sampled from three topographic zones: plateau, hillslope and lowland. In each zone, a 25 m transect was

established prior to the first sampling. On each sampling date, eight chambers were installed at randomly selected locations along each transect. Chambers were installed 5 min before sampling and were removed after each collection. Chambers consisted of 32 cm-diameter PVC rings and caps (volume approximately 5.6 L). The rings were inserted 2 cm into the soil. The caps contained small flexible membranes that maintained similar pressure inside and outside the chamber during sampling. Gas samples were collected manually with 60-mL polypropylene syringes before closing the chamber and after 5, 10, 15 and 20 min. Immediately after sampling, the gas was transferred to evacuated serum bottles capped with Teflon septa. Soil and air temperature were collected during the incubation and chamber volumes were measured manually after each measurement.

To test whether installation of chambers before collections altered gas fluxes compared with fixed chambers, we conducted a one-time experiment to compare CO_2 and CH_4 fluxes from fixed and moveable chambers during the dry season of 2008. We installed four fixed chambers at random locations along the 25 m transect in the hillslope three days before sampling. We then compared fluxes in these four chambers with those measured in four adjacent chambers installed 5 min before sampling. Fluxes from all chambers were collected between 11 am and 2 pm on three successive days. We tested for differences between fixed moveable chambers using *t*-tests ($\alpha = 0.05$).

All gas samples were transported to the laboratory at the University of São Paulo in Piracicaba where they were analyzed within 30 days on a Shimadzu GC17A gas chromatograph equipped with a flame ionization detector. A methanizer module was used to covert CO_2 to CH_4 (Shimadzu Scientific Instruments). Detection limits were 4.63 ppm for CO_2 and 0.002 ppm for CH_4. Measurement precision was ± 0.11 ppm for CO_2 and ± 0.003 ppm for CH_4. CO_2 and CH_4 fluxes were calculated from linear regressions of gas concentration and sampling time. We used liner regression to examine relationships between gas fluxes and cumulative rainfall amounts in the period prior to sampling.

Stream–atmosphere fluxes of CO_2 and CH_4

Fluxes of CO_2 were measured directly from the stream surface using a dynamic plexiglass floating chamber (0.125 m^2, 13.5 L) connected to a LICOR Model 820 infra-red gas analyzer (Sebacher and Harriss 1982). The chamber was outfitted with a pump to circulate air between chamber and gas analyzer. Concentrations of CO_2 were recorded every second over 6 min. Concentrations were recorded on a portable computer connected to the IRGA. Four to five separate measurements were performed on each sampling date. Stream surface flux measurements were made approximately monthly during the dry season and approximately biweekly during the rainy season. Flux rates were calculated from the linear rate of change of CO_2 concentrations.

The same chamber was transformed into a static chamber to determine stream-atmosphere CH_4 fluxes by replacing the re-circulating loop with a vent to allow sampling with a syringe. Separate measurements were made for CH_4 flux. Gas was collected in 60-mL syringes after 1, 2, 3, 4 and 5 min. Samples were immediately transferred to evacuated serum bottles, transported to the University of São Paulo and analyzed within 30 days as described above.

Streamwater pCO_2 concentrations and streamwater C export

Streamwater pH was measured in situ with a calibrated Orion 250A + meter. The pCO_2 of streamwater was determined by equilibrating streamwater with an evacuated headspace in sealed containers. At each collection date, triplicate 1 L bottles were filled with streamwater with no headspace. A CO_2-free headspace was created by introduction of 60 mL of N_2 gas through a 2-port silicone stopper, while withdrawing with another syringe 60 mL of sample from the bottom of the bottle. The bottle was shaken for 3 min and then left to incubate in the stream for 15 min, after which the headspace gas was extracted while initially removed sample water was injected back into the bottom of the bottle. Headspace sample withdrawn with syringes were immediately injected into evacuated serum bottles. The CO_2 concentration was determined on the gas chromatograph described above.

Concentrations of coarse particulate organic C (POC) were measured by passing a known volume of water through a 63 μm sieve, drying and weighing the collected material and analyzing it for total C content. Volumes varied according to the visual analysis of the amount of material that was retained

in the sieve, in order to assure that enough particles were collected for further processing. Fine POC was measured by filtering the sieved water through pre-weighed, ashed 0.7 μm GF/F filters, drying and analyzing for total C. Total C was analyzed on a Fisons EA 1110 CHNS connected to a Finnigan Delta Plus mass spectrometer. POC was sampled approximately monthly during the dry season and approximately bi-weekly during the rainy season.

Samples for DOC and DIC were collected concurrently with POC. Triplicate DOC samples were collected by filtering streamwater through ashed GF/F filters into 25 mL glass vials equipped with Teflon lids and preserved in the field with 25 μL of 300 mM $HgCl_2$. Another sample was collected for DIC, filtered through a 0.45 μm PALL Supor-450 membrane filter into 60 mL HDPE bottles and preserved with 100 mg of thymol. DOC and DIC were determined separately in a Shimadzu total carbon analyzer (Model TOC-VCPH). DOC was measured on $HgCl_2$ preserved samples after the extraction of DIC by acidification and sparging. DIC was measured directly as DIC in the other sample preserved with thymol. The limits of detection and sensitivities of the methods are 0.06 ppm and 0.09 for DOC, respectively, and 0.036 ppm and 0.43 for DIC.

We estimated CO_2 outgassing fluxes from the watershed in three ways: (1) direct measurement from the stream surface flux chamber, (2) calculating potential outgassing assuming that measured elevated streamwater pCO_2 would equilibrate with the atmosphere downstream, and (3) estimating outgassing from bypassing groundwater based on the modeled bypass flow and estimated groundwater pCO_2.

To estimate the potential outgassing by equilibration of streamwater, we calculated the mass of dissolved CO_2 based on measured pCO_2, pH and temperature (Skirrow 1975; Szaran 1998) under stream conditions. To estimate the potential CO_2 outgassing in the flow of bypassing groundwater we assumed a groundwater pCO_2 of 60,000 μatm, which is in the upper portion of the range of pCO_2 measured in Amazon groundwater (Richey et al. 2009).

We calculated fluvial export of DOC, DIC and fine and coarse particulate organic C (FPOC, CPOC) from stream discharge and streamwater concentrations by determining mid-points between sampling dates, assigning measured concentrations to the measured total discharge for each period and summing fluxes for each period. We also estimated DIC and DOC exported in bypassing groundwater from modeled groundwater discharge and mean concentrations of DIC (26.1 ± 23.3 mg C L^{-1}) and DOC (1.47 ± 0.39 mg C L^{-1}) measured approximately monthly in 9 groundwater wells (3 each on plateau, hillslope and lowland). Groundwater wells were installed by hand at different depths depending on their position on the plateau-lowland transect. All wells were screened 1.5 m into the groundwater. Samples were obtained by lowering a PVC bailer connected to a measuring tape, after removing all the water and waiting for the wells to fill again. We assumed no POC was transported in bypassing groundwater.

Results

Precipitation in 2007 totaled 1,641 mm and was lower than the 1987–2007 mean of 1,905 mm (Fig. 2). The greatest difference between measured

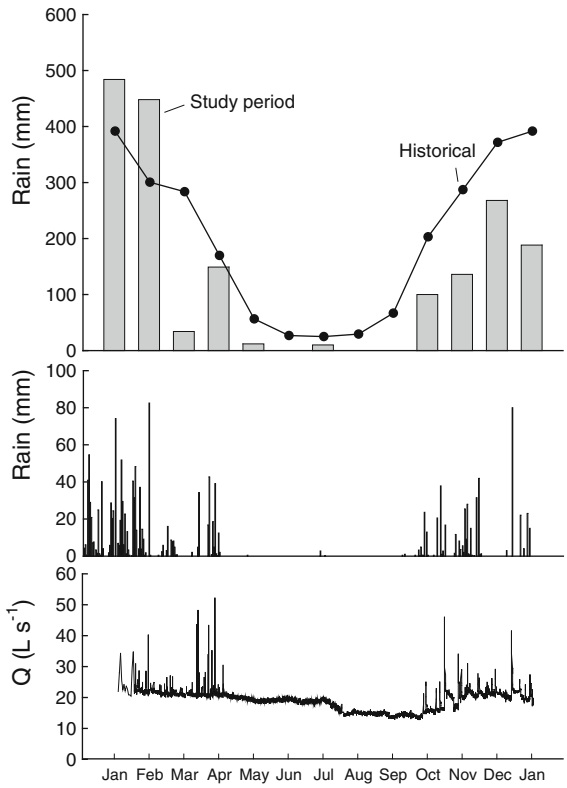

Fig. 2 Recent historical (1987–2007) monthly precipitation and precipitation at Tanguro Ranch during the January 2007 to January 2008 study period (*top*), daily rainfall (*middle*) and stream discharge (*bottom*)

and historical mean precipitation occurred in March 2007 when precipitation of 34 mm was lower than the longer term average of 250 mm. Stream discharge ranged from 13 to 52 L s^{-1} and was largely aseasonal, but stormflow was nonexistent between April and October (Fig. 2). Mean dry season discharge was 16.6 ± 2.2 L s^{-1} and mean rainy season discharge was 20.8 ± 2.1 L s^{-1}. Baseflow accounted for 96% of annual streamflow. Mean stream discharge measured on five dates increased from 4.7 L s^{-1} at the upstream point to 22.7 L s^{-1} at the downstream point and indicated a mean increase of 0·18.0 L s^{-1} km^{-1}.

Total stream discharge was 60 mm y^{-1}. This compared with modeled evapotranspiration of 1,149 mm y^{-1} and yielded a total watershed runoff of 492 mm y^{-1} (Table 1). This indicated that 432 mm y^{-1} bypassed the stream gauging point as deep groundwater that discharged downstream in the watershed (Table 1).

There were no differences in CO_2 or CH_4 fluxes between fixed and movable soil chambers ($p < 0.05$). Soil CO_2 fluxes were highest during the wettest periods of the year, February to March and from December to January (Fig. 3). Soil CO_2 fluxes were

Table 1 Water balance at Tanguro Ranch

	Rain mm	Evapotranspiration	Watershed runoff	Stream runoff
Average (1987–2007)	**1,905**	1,149	571	–
15 January 2007–January 2008	**1,149**	1,149	492	**60**

Measured values are indicated in *bold*

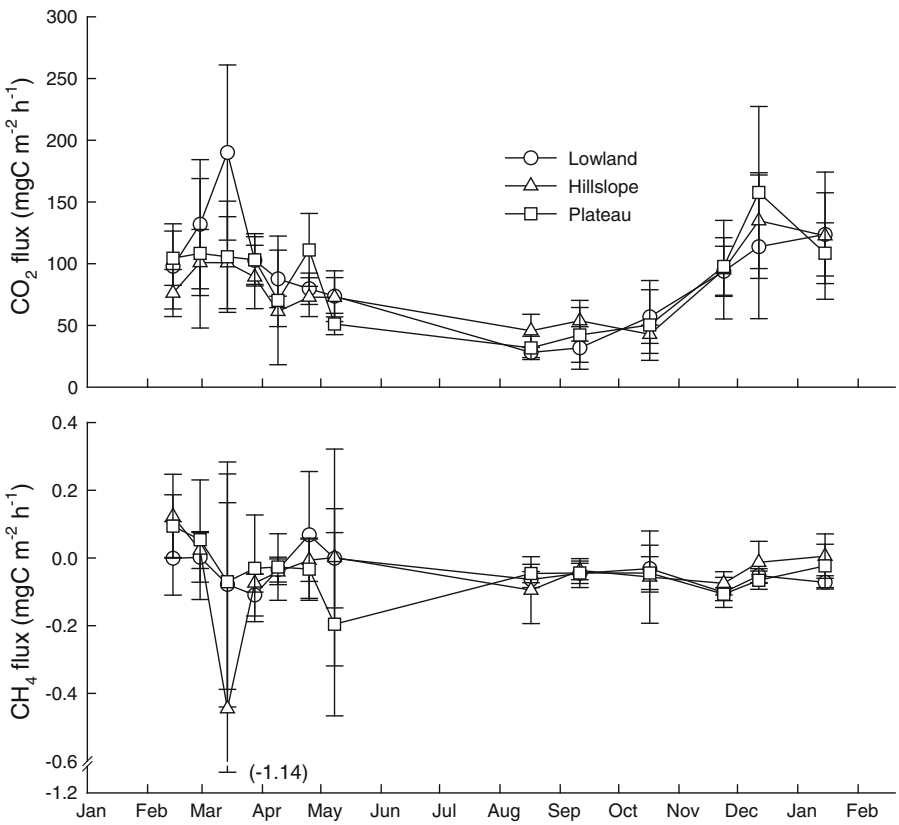

Fig. 3 Measured soil CO_2 and CH_4 fluxes from lowland, hillslope and plateau

Table 2 Comparison of soil CO_2 and CH_4 fluxes from fixed and movable chambers

	Day	Fixed chamber	Movable chamber	*t*-Value	*p*<
Flux (mg C m^{-2} h^{-1})	1	61 ± 18	63 ± 18	−0.137	0.895
	2	37 ± 3	46 ± 12	−1.440	0.197
	3	54 ± 4	53 ± 8	0.11	0.91
	Mean	50 ± 15	54 ± 15		
Flux (mg C m^{-2} h^{-1})	1	−0.11 ± 0.07	−0.03 ± 0.05	−1.450	0.2
	2	−0.08 ± 0.05	−0.05 ± 0.02	−1.990	0.093
	3	−0.07 ± 0.01	−0.07 ± 0.02	0.065	0.95
	Mean	−0.09 ± 0.04	−0.05 ± 0.04		

positively correlated with accumulated precipitation in the 15 days before sampling ($r^2 = 0.81$, $p < 0.0003$). Soil CO_2 fluxes were not correlated with soil or air temperature. Soil CO_2 fluxes did not differ among plateau, hillslope and lowland. (Table 2).

Soils consumed small amounts of atmospheric CH_4 for most of the year (Fig. 3). Soils were a net source of CH_4 to the atmosphere during the wet season in February 2007 but were net sinks of CH_4 during the drier-than-average period during March 2007. Soil CH_4 fluxes did not differ among plateau, hillslope and lowland.

Annual soil CO_2 emissions ranged from 683 ± 129 g C m^{-2} y^{-1} on the hillslope to 721 ± 160 g C m^{-2} y^{-1} in the lowland (Table 3). Annual CO_2 fluxes did not differ among plateau, hillslope and lowland. Soils from all three locations were CH_4 sinks that ranged from −0.7 ± 1.2 g C m^{-2} y^{-1} on the plateau to-0.3 ± 0.7 g C m^{-2} y^{-1} in the lowland (Table 3). Annual CH_4 fluxes did not differ among lowland, hillslope and plateau. At the scale of the watershed, the plateau was the dominant source of soil CO_2 flux (Table 3) because of the large areal extent of the plateau (99.7% of total watershed land area). Soil CO_2 emission from the watershed was 696 ± 147 g C m^{-2} y^{-1} (Table 3). The plateau was also the dominant sink for CH_4 in the watershed, but CH_4 fluxes were 3 orders of magnitude lower than CO_2 fluxes and played an insignificant role in the total basin gaseous C emissions from soils (Table 3).

Streamwater pH was 4.43 ± 0.39. The stream had high pCO_2 concentrations of 6,491–14,976 µatm that indicated significant supersaturation (15–45 times) of stream water relative to the atmosphere (Fig. 4). Stream pCO_2 and stream surface fluxes of CO_2 to the atmosphere were variable but were generally higher in the wet season (Fig. 4). The annual average rate of CO_2 from the stream surface was 5,994 ± 677 g C m^{-2} y^{-1} (Table 4), nearly 10 times greater than the flux rate per unit area from soils. Stream pCH_4 and stream surface CH_4 fluxes showed little consistent seasonal variation (Fig. 4). Annual flux of CH_4 from the stream surface was 987 ± 221 g C m^{-2} y^{-1} (Table 4) and was consistent source of CH_4 to the atmosphere in contrast to soils, which consumed CH_4 on an annual basis. Because the area of the stream surface was small compared with land area, fluxes of both CO_2 and CH_4 extrapolated for the entire basin were low. Annual measured outgassing of CO_2 was 0.44 ± 0.05 g C m^{-2} y^{-1} and outgassing of CH_4 was 0.07 ± 0.02 g C m^{-2} y^{-1} (Table 4).

We estimated that a much larger amount of CO_2 (10.39 g C m^{-2} y^{-1}) was exported from the watershed by outgassing of groundwater that bypassed our stream gauge. Calculation of potential CO_2 outgassed to the atmosphere based on equilibration of measured stream pCO_2 gave a much lower estimate of 0.19 g C m^{-2} y^{-1} (Table 4).

DIC concentrations (here and therein not accounting gaseous CO_2) in streamwater were greater than DOC concentrations throughout the year and both DIC and DOC showed no consistent seasonal pattern (Fig. 5). Concentrations of FPOC and CPOC were two orders of magnitude smaller. FPOC concentrations were highest immediately after the first rains of the rainy season (Fig. 5).

DIC and DOC were the largest components of measured fluvial C export from the watershed. Measured DIC export per unit area of watershed

Table 3 Basin area and annual CO_2 and CH_4 fluxes from soils at three topographic positions

	Plateau	Hillslope	Lowland	Total watershed
Area (ha)	1,315	2.5	1.2	1,319
CO_2 flux (g C m^{-2} y^{-1})	695 ± 145	683 ± 129	721 ± 160	696 ± 145
Total watershed CO_2 flux (Mg C y^{-1})	9,132 ± 1,910	17.06 ± 3.23	8.9 ± 1.97	9,158 ± 1,915
CO_2 flux per m^{-2} of watershed (g C m^{-2} y^{-1})	694 ± 146	1.3 ± 0.3	0.7 ± 0.2	696 ± 145
CH_4 flux (g C m^{-2} y^{-1})	−0.7 ± 1.2	−0.3 ± 1.5	−0.3 ± 0.7	−0.7 ± 1.2
Total watershed CH_4 flux (Mg C y^{-1})	−9.2 ± 15.8	−0.008 ± 0.038	−0.0035 ± 0.009	−9.21 ± 15.8
CH_4 flux per m^{-2} of watershed (g C m^{-2} $y^{-1)}$	695 ± 146	1.3 ± 0.3	0.7 ± 0.2	696 ± 147

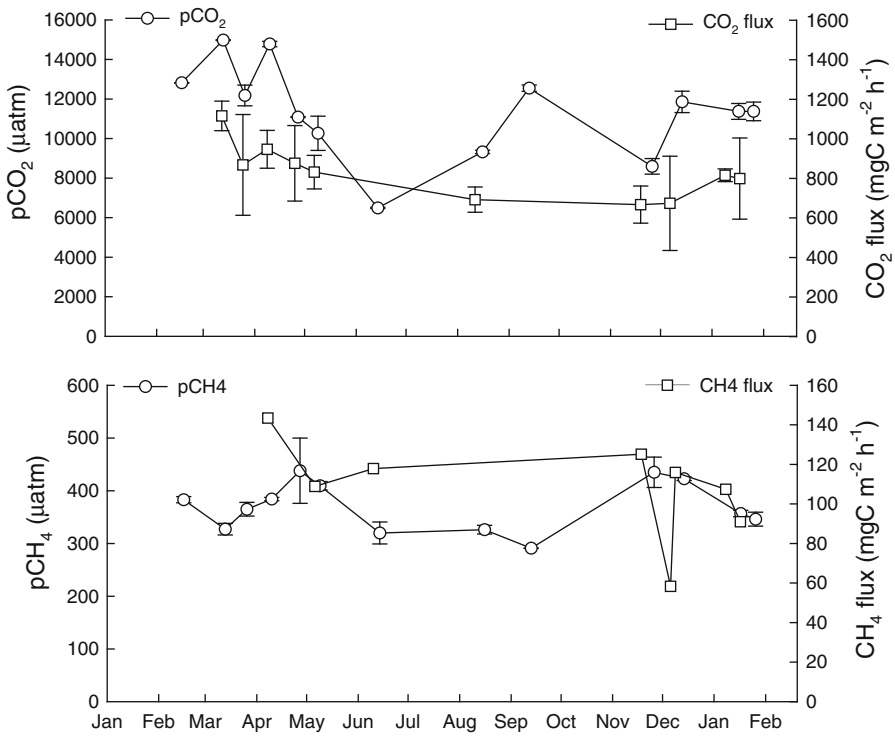

Fig. 4 Concentrations of pCO_2 and CO_2 fluxes (*top*) and $pCH4$ and CH_4 fluxes (*bottom*)

Table 4 Hourly and annual fluxes of CO_2 and CH_4 from the stream channel and total fluxes from the stream channel per unit are of watershed

Flux	Units	Method	CO_2	CH_4
Mean hourly flux from channel	mg C m^{-2} h^{-1}	Measured	766 ± 261	108 ± 25
Annual flux from channel	g C m^{-2} y^{-1}	Measured	5,994 ± 677	987 ± 221
Total annual flux from channel	kg C y^{-1}	Measured	5,814 ± 647	957 ± 214
Total annual flux from channel	g C m^{-2} watershed y^{-1}	Measured	0.44 ± 0.05	0.07 ± 0.02
Total annual flux assuming equilibration of stream CO_2	g C m^{-2} watershed y^{-1}	Estimated	0.19	
Total annual flux in bypassing groundwater per m^2 of watershed	g C m^{-2} watershed y^{-1}	Estimated	10.39	

The area of the stream channel at base flow was 970 m^2. Annual flux in bypassing groundwater was estimated assuming groundwater pCO_2 of 60,000 µatm (Richey et al. 2009)

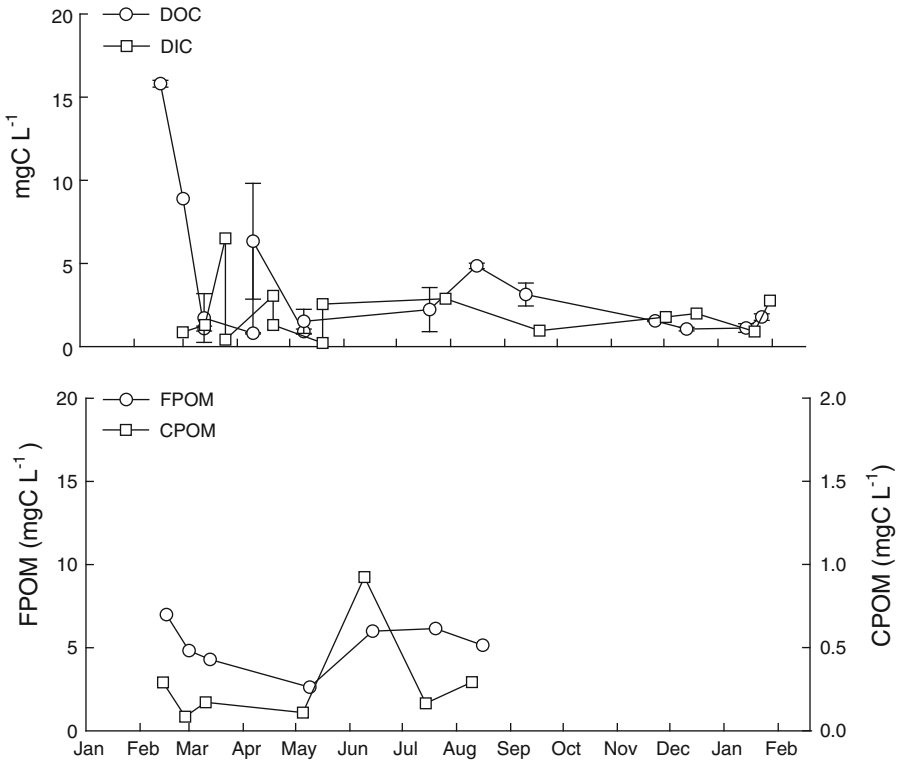

Fig. 5 Concentrations of dissolved inorganic C and dissolved organic C (*top*) and concentrations of C in fine particulate organic C and coarse particulate organic C (*bottom*)

was 0.78 ± 0.04 g C m^{-2} y^{-1}, measured DOC export was 0.16 g C m^{-2} y^{-1} and measured FPOC and CPOC export were 0.001 g C m^{-2} y^{-1} or less (Table 5).

We estimated that a larger amount of DIC (11.29 g C m^{-2} y^{-1}) and DOC (0.64 g C m^{-2} y^{-1}) were exported in bypassing groundwater compared with export directly measured in streamwater (Table 5). The total estimated streamwater plus groundwater export of DIC and DOC of 12.87 g C m^{-2} y^{-1} was of similar magnitude to the total estimated streamwater plus groundwater export of CO_2 by outgassing of 10.83 g C m^{-2} y^{-1}.

Discussion

Soil emissions

The rates of soil CO_2 emission in all three topographic zones were lower than the range of rates of 140–240 mg C m^{-2} h^{-1} measured in other studies of lowland moist Amazon forest (Meir et al. 1996; Chambers et al. 2004; Garcia-Montiel et al. 2004; Davidson et al. 2008). Our soil CO_2 fluxes were correlated with the total accumulated precipitation before flux measurements, indicating that soil moisture is an important controlling factor in this process, a result similar to other studies in the Amazon (Saleska et al. 2003; Salimon et al. 2004; Vourlitis et al. 2004). The soil CO_2 fluxes we measured were more similar to the approximately 130 mg C m^{-2} h^{-1} measured in Brazilian cerrado (savanna) (Meir et al. 1996) and suggests that the long dry season experienced by the forest at Tanguro Ranch results in rates of annual soil CO_2 emissions that more closely resemble those from cerrado.

Rates of soil CH_4 consumption were also generally lower than those found in other studies of moist tropical forest (Keller et al. 2005; Steudler et al. 1996; Davidson et al. 2008). The seasonality we observed of soils acting as small sinks for CH_4 in the dry season and small sources during the rainy season was also similar to other tropical forest locations where methanotropy dominates over methanogenesis during the dry season (Keller and Reiners 1994; Steudler et al.1996; Keller et al. 2005; Davidson et al. 2008). In our site, soil bulk density was low and soils

Table 5 Fluvial export of dissolved and particulate inorganic and organic C

Flux	Measured		Estimated in bypassing groundwater	Total
	kg C y^{-1}	g C m^{-2} watershed y^{-1}	g C m^{-2} watershed y^{-1}	g C m^{-2} watershed y^{-1}
DIC (ionic forms only)	10,339 ± 514	0.78 ± 0.04	11.29 ± 10.01	12.07 ± 10.05
DOC	2,122 ± 360	0.16 ± 0.03	0.64 ± 0.17	0.80 ± 0.20
Fine POC	16.24 ± 8.6	0.001 ± 0.001	0	0.001 ± 0.001
Coarse POC	0.78 ± 0.03	0.00006 ± 0.000002	0	0.00006 ± 0.00002

were consistently well drained and these conditions were consistent with methane consumption during most of the year.

Stream-atmosphere fluxes

Many surface waters of the Amazon are supersaturated in CO_2 in relation to the atmosphere (Richey et al. 2002, 2009), with the highest dissolved CO_2 concentrations typically found in headwater streams (Johnson et al. 2008; Razera et al. 2008; Rosa 2007). The absence of carbonate-rich rocks in the highly weathered landscape of the Brazilian Shield and isotopic signatures of CO_2 indicate transfer of CO_2 from deep soils to streams via lateral movement of groundwater (Johnson et al. 2008). Johnson et al. (2008) demonstrated that headwater streams in the Amazon receive large amounts of CO_2 from adjacent land via seepage water, which evades to the atmosphere within several hundred meters of its emergence in stream channels. This phenomenon has been observed in a variety of small streams and lakes that receive inputs of groundwater from surrounding terrestrial landscapes (Kling et al. 1991; Jones and Mulholland 1998a, b; Dawson et al. 2002; Hope et al. 2004).

Our results supported the idea that headwater streams were landscape hotspots for CO_2 emission. Our measured stream CO_2 fluxes of 674–1,116 mg C m^{-2} h^{-1} (15.6–25.8 μmol CO_2 m^{-2} s^{-1}) were higher than most of the small number of other direct measurements of CO_2 flux from stream surfaces. In a survey of 21 streams and rivers of third to fifth orders in Rondônia, Razera et al. (2008) found that fluxes averaged 5.6 ± 3.1 μmol CO_2 m^{-2} s^{-1}. Higher flux rates from streams in the Amazon were found only by Rosa (2007) in first order streams at the eastern Pará, where values reached up to 53 μmol CO_2 m^{-2} s^{-1}.

Our results also indicated that headwater streams can also serve as hotspots for CH_4 emission. Although there are no other reported measurement of these fluxes from headwater streams of the Amazon, our measured stream CH_4 fluxes of 108 ± 25 mg C m^{-2} h^{-1} were comparable to fluxes of 25–250 mg C m^{-2} h^{-1} measured from open water areas and streams draining boreal peatland (Billett and Moore 2008). Our fluxes were high compared with other studies of CH_4 emissions from Amazon aquatic environments and wetlands in general. For example, Devol et al. (1988) found emissions from lakes, macrophyte beds and flooded forest the central floodplain ranged from 0.04 to 32 mg C m^{-2} h^{-1}. Estimates of CH_4 fluxes more generally from wetland environments range from 5 to 10 mg C m^{-2} h^{-1} (Bartlett and Harriss 1993).

Our measurements were made with chambers that can disrupt eddies and air movement near the water surface, cause additional water turbulence in flowing water or inhibit gas production as gas concentrations build up in the chamber (Lapitan et al. 1999; Matthews et al. 2003). Although these effects may be relatively minor (Park et al. 2010), chambers can overestimate fluxes compared with direct measurements of transfer coefficients (k) using SF_6 by a factor of about two, particularly in calm environments such as those under forest canopies (Matthews et al. 2003). In stream environments that spanned open and flowing water, Billett and Moore (2008) showed that chambers generally underestimated CO_2 fluxes compared with estimation from winds speed in open water environments but overestimated CO_2 fluxes in flowing waters. Our sampling location was flowing but with little turbulence, suggesting that our chamber measurements may have slightly overestimated fluxes.

Our directly-measured chamber-based fluxes of 0.44 g C m^{-2} y^{-1} captured only a small portion of the total watershed CO_2 outgassing of 10.83 g C m^{-2} y^{-1} that we estimated by including CO_2

carried by the relatively large volume of water that was exported from the watershed as discharge of groundwater downstream of our stream gauging and sampling point. This discrepancy points out the importance of including these deep groundwater flows in estimates of watershed water balances in very small watersheds, particularly on the deep and highly permeable soils in areas of low relief that make up much of the central lowland Amazon. This larger estimate of export assumed that all groundwater had a pCO_2 of 60,000 μatm, which is at the higher end of the range measured in Amazon groundwater (Richey et al. 2009) and provides a reasonable upper bound on watershed CO_2 outgassing contained in bypassing groundwater.

Because pCO_2 declines rapidly downstream of groundwater discharge seeps (Johnson et al. 2008) it is possible that our direct measurements of CO_2 outgassing underestimated this flux because measurements were made from streamwater rather than from springs directly emerging from the ground. The Tanguro stream did not have a well-defined spring source but was formed by relatively uniform entry of groundwater by bank seepage. Our chamber therefore sampled a mix of recently-emerged groundwater (presumably high in CO_2) and water that emerged farther upstream (from which most CO_2 had already evaded) that was likely typical of the stream reach. Sampling at the upstream-most seepage area would not have provided an unambiguous estimate of the pCO_2 in emerging groundwater because it would have been influenced by very low flow rates that would have allowed some equilibration near the soil surface.

This study and others suggest that the CO_2 outgassing fluxes for small Amazon streams is in the range of 2–40 g C m^{-2} y^{-1} (0.02–0.4 Mg C ha y^{-1}). Our estimated CO_2 outgassing flux of 10.8 g C m^{-2} y^{-1} was less than the 40 g C m^{-2} y^{-1} (0.4 Mg C ha^{-1} y^{-1}) measured from small watersheds in Mato Grosso by Johnson et al. (2008) and the approximately 50 g C m^2 y^{-1} (0.5 Mg C ha^{-1} y^{-1}) originating from soils and respiration of terrestrial DOC that was estimated to be outgassed from large central Amazonian rivers by Richey et al. (2002). However, our fluxes were larger than the CO_2 outgassing fluxes of 2–4 g C m^{-2} y^{-1} from three streams in the eastern Amazon by Davidson et al. (in press). The differences in these estimates likely result from several factors. Lower rainfall at Tanguro compared with the other sites, particularly during our year of measurement, resulted in a lower estimated flux compared with streams sampled in Mato Grosso and Pará. This is supported by a model estimate of higher stream CO_2 outgassing fluxes from the regions of the Amazon with highest rainfall (Richey et al. 2009). Our results also suggest that streams in which most water arrives as deep groundwater flow have greater potential to export CO_2 through the outgassing pathway because of long water contact time with soils. While there is no solid evidence that CO_2 concentrations in deep soils differ between forest and other land uses (Davidson and Trumbore 1995), the more than 15-fold increase in the volume of water that reaches streams by quick (surface or near-surface) flowpaths when forest is converted to pasture (Moraes et al. 2006; Germer et al. 2009) will likely reduce watershed export by CO_2 outgassing because more streamwater will be derived from water that remained in greater contact with the atmosphere. Estimates based on direct measurements of flux or pCO_2 in streamwater also depend on the extent to which CO_2 had evaded from emerging groundwater at the location where samples were collected. That effect was minimal in the spring sampled by Johnson et al. (2008) but potentially important in the streams sampled by Davidson et al. (in press) and in our study, although in our case this had much less of an effect because our estimate of CO_2 released from bypassing groundwater assumed no equivalent downstream loss for 88% of total water export that left the watershed via this pathway.

Fluvial exports

Our measured streamwater DIC concentrations (6–20 mg C l^{-1}) and export were similar to those reported for other small Amazon headwater streams (Johnson et al. 2006a, 2008) and our measured DIC exported by streamwater was substantially less than CO_2 exported directly from the stream by outgassing. However, total estimated watershed export of DIC (12.1 g C m^{-2} y^{-1}) was comparable in magnitude to total watershed export by outgassing of CO_2 (10.83 g C m^{-2} y^{-1}). This was driven largely by a high average concentration of DIC in groundwater (26.1 mg C l^{-1}) and suggests that export of DIC can rival the CO_2 outgassing flux.

Watershed export of DOC of 0.8 g C m^{-2} y^{-1} was low and substantially less than measured at some other

locations. Our streamwater DOC concentrations ranged from 0.3 to 15 mg C l^{-1} but were generally <1 mg C L^{-1}. These were comparable to DOC in the Mato Grosso stream measured by Johnson et al. (2006b, 2008) but much lower than the range of 9–27 mg C L^{-1} reported in a forest watershed in a DOC-rich blackwater region from the central Amazon (Waterloo et al. 2006). In general, Amazonian blackwater streams and rivers own their color to high amounts of DOC, whereas clear waters show much smaller concentrations of these forms of dissolved carbon (McClain and Richey 1996). Our total estimated watershed DOC export of 0.80 g C m^{-2} y^{-1} was much lower than the range of 13–28 g C m^{-2} y^{-1} exported from that stream as DOC in two different years (Waterloo et al. 2006). This substantial difference in the role of DOC as a pathway for ecosystem C loss between these two different Amazon environments almost certainly arises from combination of deep, well-drained soils at Tanguro and deep groundwater flowpaths that result in very low groundwater DOC concentrations.

Fluvial export of POC was a very minor component of watershed C export. We measured fluvial export of particulate C of <0.001 g C m^{-2} y^{-1} that was extremely low compared with estimated of particulate C loss of 0.9–1.8 g C m^{-2} y^{-1} from other Amazon headwater streams (Waterloo et al. 2006; Johnson et al. 2008). POC export from the Tanguro stream is likely strongly influenced by the very low energy environment and the near-complete absence of stormflow (4% of annual streamflow).

Comparison with terrestrial C fluxes

Richey et al. (2002) postulated that CO_2 derived from respiration of roots and organic matter in the soils of terrestrial ecosystems and the respiration of terrestrially-derived DOC could account for outgassing from large rivers of about 0.5 Mg C ha^{-1} y^{-1} from a nearly 2 million km^2 area of the central Amazon. The results of this study and the small number of studies that directly measured terrestrial-aquatic transfers of C in small forested watersheds suggest that the export of terrestrially-derived C through small streams is likely to be similar to or somewhat smaller than this original estimate. However, these stream-associated watershed C losses can be significant compared with net annual ecosystem C storage or loss in the range of ±150 g C m^2 y^{-1} (±1.5 Mg ha^{-1} y^{-1}) estimated for Amazon forest (Saleska et al. 2003; Miller et al. 2004; Vourlitis et al. 2004; Phillips et al. 2009). There is considerable variation in the magnitude of this transfer in different rainfall and soil environments. Estimates of stream-associated CO_2 outgassing fluxes would be improved by more widespread measurements of pCO_2 and DIC in groundwater and groundwater springs or seepage.

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