

## Carbon dioxide and methane emissions and the carbon budget of a 10-year old tropical reservoir (Petit Saut, French Guiana)

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[1] The emissions of carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) from the Petit Saut hydroelectric reservoir (Sinnamary River, French Guiana) to the atmosphere were quantified for 10 years since impounding in 1994. Diffusive emissions from the reservoir surface were computed from direct flux measurements in 1994, 1995, and 2003 and from surface concentrations monitoring. Bubbling emissions, which occur only at water depths lower than 10 m, were interpolated from funnel measurements in 1994, 1997, and 2003. Degassing at the outlet of the dam downstream of the turbines was calculated from the difference in gas concentrations upstream and downstream of the dam and the turbined discharge. Diffusive emissions from the Sinnamary tidal river and estuary were quantified from direct flux measurements in 2003 and concentrations monitoring. Total carbon emissions were  $0.37 \pm 0.01$  Mt yr<sup>-1</sup> C (CO<sub>2</sub> emissions,  $0.30 \pm 0.02$ ; CH<sub>4</sub> emissions,  $0.07 \pm 0.01$ ) the first 3 years after impounding (1994–1996) and then decreased to  $0.12 \pm 0.01$  Mt yr<sup>-1</sup> C (CO<sub>2</sub>,  $0.10 \pm 0.01$ ; CH<sub>4</sub>,  $0.016 \pm 0.006$ ) since 2000. On average over the 10 years, 61% of the CO<sub>2</sub> emissions occurred by diffusion from the reservoir surface, 31% from the estuary, 7% by degassing at the outlet of the dam, and a negligible fraction by bubbling. CH<sub>4</sub> diffusion and bubbling from the reservoir surface were predominant (40% and 44%, respectively) only the first year after impounding. Since 1995, degassing at an aerating weir downstream of the turbines has become the major pathway for CH<sub>4</sub> emissions, reaching 70% of the total CH<sub>4</sub> flux. In 2003, river carbon inputs were balanced by carbon outputs to the ocean and were about 3 times lower than the atmospheric flux, which suggests that 10 years after impounding, the flooded terrestrial carbon is still the predominant contributor to the gaseous emissions. In 10 years, about 22% of the 10 Mt C flooded was lost to the atmosphere. Our results confirm the significance of greenhouse gas emissions from tropical reservoir but stress the importance of: (1) considering all the gas pathways upstream and downstream of the dams and (2) taking into account the reservoir age when upscaling emissions rates at the global scale.

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### 1. Introduction

[2] The atmospheric concentrations of the two major greenhouse gases, carbon dioxide and methane, have almost continuously risen since the beginning of the last century [Raynaud *et al.*, 1993], meaning that their sources at the

Earth surface have become much larger than their sinks [Intergovernmental Panel on Climate Change, 2001]. It is well established that fossil fuel combustion is by far the major contributor to the CO<sub>2</sub> increase in the atmosphere. By contrast, the increase in CH<sub>4</sub> is still partially understood today, since it is the cumulative result of numerous impacts

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of man activities that increase emissions from the Earth surface: rice cultivation, coal and gas mining, ruminants breeding, biomass burning and landfill spills [Cicerone and Oremland, 1988].

[3] In the last decade, artificial reservoirs have also been identified as significant CO<sub>2</sub> and CH<sub>4</sub> sources to the atmosphere [Rudd et al., 1993; Rosa and Schaeffer, 1994; Duchemin et al., 1995; Galy-Lacaux et al., 1997, 1999; Delmas et al., 2001; Rosa et al., 2003; Soumis et al., 2004; Tremblay et al., 2004]. In the tropics, high temperatures and the flooding of large amounts of biomass, including primary forest, result to intense CO<sub>2</sub> and CH<sub>4</sub> production at reservoir bottoms [Fearnside, 1995; Galy-Lacaux et al., 1997, 1999; Delmas et al., 2001; Rosa et al., 2003]. Tropical reservoirs emit CH<sub>4</sub> at rates often similar to Amazonian wetlands [Richey et al., 1988]. Saint Louis et al. [2000] have estimated that world artificial reservoirs could emit 64 Mt yr<sup>-1</sup> CH<sub>4</sub>, 90% of the emissions occurring in the tropics. If true, this would increase previous CH<sub>4</sub> emission estimations from the Earth surface by 15 to 20% [Cicerone and Oremland, 1988; Stern and Kaufman, 1996]. This estimate is, however, based on a limited amount of measurements. Although they possibly play a significant role in the global CH<sub>4</sub> budget, few tropical reservoirs have been yet investigated in Panama [Keller and Stallard, 1994], in Brazil [Matvienko et al., 2000; Rosa et al., 2003] and in French Guiana [Galy-Lacaux et al., 1997, 1999]. Furthermore, the succession of wet and dry seasons in the tropics induces large seasonal changes in gaseous emissions from reservoirs. These temporal variations on a single system are generally poorly constrained, although they might be very significant.

[4] The major process responsible for the high CO<sub>2</sub> and CH<sub>4</sub> emissions from young reservoirs is the microbial decomposition of the flooded terrestrial organic matter (OM). This flooded OM is composed of soils, litters, trunks as well as leaves that generally fall into the water few months after flooding. Gaseous emissions are maximum the first 2 to 3 years after impounding and then slowly decrease with time [Kelly et al., 1997; Galy-Lacaux et al., 1999; Saint Louis et al., 2000; Rosa et al., 2003]. This shows that a more labile fraction of the flooded terrestrial OM is first rapidly lost, followed by a progressive decrease in quantity and bioavailability of the OM remaining at reservoirs bottoms. In addition, in old reservoirs, other sources of OM (river input and aquatic primary production) become significant for decomposition in reservoirs. Furthermore, the installation of an active population of methanotropic bacteria may take some time and CH<sub>4</sub> oxidation may become significant only several months after impounding [Galy-Lacaux et al., 1997; Dumestre et al., 1999]. Superimposed on this long-term trend, gaseous emissions from hydroelectric reservoirs also show very large temporal variations with meteorological conditions (wind and rainfall) that change gas exchange rates and with river discharge that modifies the residence time of the waters. Long-term monitoring of CO<sub>2</sub> and CH<sub>4</sub> concentrations and fluxes with a high frequency are necessary to apprehend this variability.

[5] Among the few tropical reservoirs investigated yet, the Petit Saut system in French Guiana, flooded in 1994

constitutes an ideal case study. CO<sub>2</sub> and CH<sub>4</sub> fluxes were studied the first three years after impounding [Galy-Lacaux et al., 1997, 1999]. In addition, monitoring of concentrations has been performed at one central station in the reservoir (vertical profiles) and at two stations in the river downstream (surface) for 10 years now. In the present paper, we present recent flux measurements performed in 2003 together with a synthesis of monitoring data. This allows us the computation of a CO<sub>2</sub> and CH<sub>4</sub> emission budget integrated over 10 years. Furthermore, a net carbon budget of the overall system is proposed and compared to the quantity of flooded organic carbon.

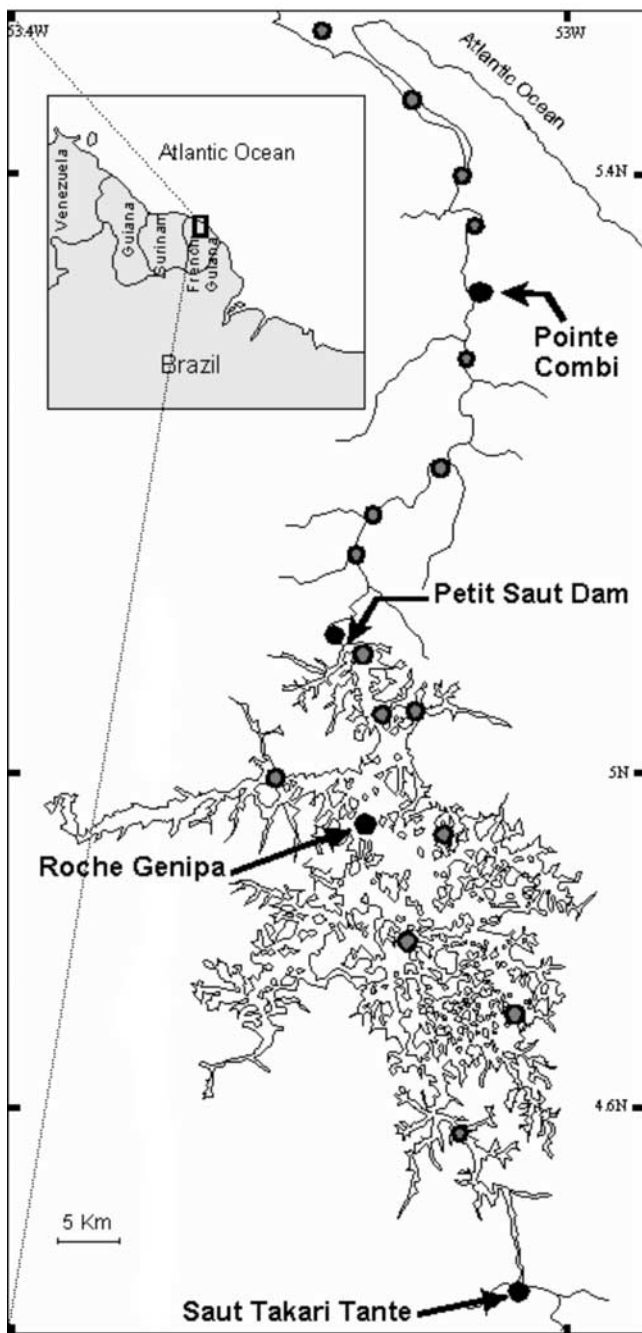
## 2. Material and Methods

### 2.1. Study Site

[6] The Petit Saut dam was constructed on the Sinnamary River in French Guiana 70 km upstream of its mouth to the Atlantic Ocean (Figure 1). The Sinnamary River has an average discharge of 260 m<sup>3</sup> s<sup>-1</sup> with important seasonal and interannual variability. The reservoir started to be filled in January 1994 and covers 80 km of the Sinnamary River course. At its maximal level of 35 m (reached in July 1995), 360 km<sup>2</sup> of uncleared tropical forest were flooded (Figure 1). Owing to the differences between high and low water levels, the average surface of the reservoir is ~300 km<sup>2</sup>. Average residence time of waters is ~5 months. The reservoir water body is highly stratified with an oxic epilimnion and an anoxic hypolimnion, separated by a quasi permanent oxicleine located around 5–7 m depths [Galy-Lacaux et al., 1999]. The reservoir outflow is located at 16 m depths. However, a 20m height submerged wall was constructed 150 m upstream of the dam in order to mix the water column and increase the oxygen concentration in the turbinated water. Downstream of the dam, an aerating weir was constructed in February 1995 in order to optimize the degassing of methane in the turbinated flow and avoid problems of hypoxia due to methane oxidation in the river [Gosse et al., 1997, 2000]. Downstream of the weir, the Sinnamary River has an average depth of 3–4 m and is influenced by the tide with an amplitude of 1.5 m. Salt intrusion starts however only at the river mouth, salinity at the limit of the coast being around 3–5. At this lower reach, the Sinnamary estuary is bordered by mangrove swamps [Fromard et al., 2004].

### 2.2. Monitoring of Carbon and Oxygen Concentrations

[7] Since impounding in 1994, oxygen, total alkalinity (TA), which at pH 5–6 is mainly HCO<sub>3</sub><sup>-</sup>, total, dissolved and particulate organic carbon (TOC, DOC and POC), and dissolved CO<sub>2</sub> and CH<sub>4</sub> were monitored at different frequency and during different periods. The station Roche Genipa (Figure 1) was chosen after 2 years as the most representative station in the reservoir for average gas concentrations. At this site, the water column (depth 30 to 35 m) was sampled monthly every 5 m from the surface to the bottom. Downstream of the reservoir, concentrations in surface waters were monitored at two stations on the Sinnamary river: one named Petit Saut dam, located



**Figure 1.** Map of the Petit Saut reservoir and the Sinnamary River and estuary in French Guiana. Black circles are reference stations where water quality was monitored at different frequencies, and CO<sub>2</sub> and CH<sub>4</sub> fluxes were measured in May and December 2003. Saut Takari Tante is located on the Sinnamary River upstream of the influence of the reservoir; Roche Genipa is 20 km upstream of the dam and is representative of the reservoir water body at its maximum depth (35 m at high waters); the Petit Saut dam station is located just downstream of the aerating weir; and Pointe Combi is located on the Sinnamary River 40 km downstream of the dam. Gray circles are additional stations where CO<sub>2</sub> and CH<sub>4</sub> fluxes were measured in May and December 2003.

0.8 km downstream of the dam and another one named Pointe Combi, located 40 km downstream of the dam (Figure 1). The measurements periods and number of data collected at each station are reported in Table 1. Basically, concentrations were measured at irregular intervals in 1994 and 1995 (from 3 to 10 times a year depending on parameters) and a more intensive monthly monitoring started in 1996 with few exceptions: no CO<sub>2</sub> data are available at station Roche Genipa for the year 2002 and TA was measured only since 1999 and not at the station 0.8 km downstream of the dam.

[8] Oxygen was measured with a polarographic electrode calibrated at 100% saturation in water saturated air. TA was measured on 1.2 μm filtrates by the Gran titration method with 0.01N HCl. CO<sub>2</sub> was measured by a headspace technique in 120 mL vials (headspace volume 50 ml) followed by analysis with a gas chromatograph equipped with a thermal conductivity detector (GC-TCD). As described by Hope *et al.* [1995] this method is the most appropriate for acid, low ionic strength, organic rich waters as the case in Petit Saut. CH<sub>4</sub> was measured using the headspace technique followed by analysis with a gas chromatograph equipped with a flame ionization detector (GC-FID). In December 2003, CO<sub>2</sub> and CH<sub>4</sub> concentrations were also measured in situ with an equilibrator [Abril *et al.*, 2005]. DOC was measured on filtrates through 0.7 μm, prerinised, glass fiber filters using an OI Analytical 1010 analyzer. The sample is acidified, purged with nitrogen to remove inorganic carbon, and oxidized with persulfate in a digestion vessel at 98°C. The resultant carbon dioxide (CO<sub>2</sub>) is measured by nondispersive infrared spectrometry. The POC concentrations were calculated as the difference between the TOC (unfiltered) and the DOC concentrations.

### 2.3. Measurements of Diffusive Gas Fluxes Using Floating Chambers

[9] Two surveys were carried out in May and December 2003, respectively during the high and low water levels in the reservoir, with turbinated discharges of 145 and 115 m<sup>3</sup> s<sup>-1</sup> respectively. Intensive direct CO<sub>2</sub> and CH<sub>4</sub> diffusive fluxes were measured at different sites: the Sinnamary River upstream and downstream of the reservoir; the estuary; the open waters and the flooded forest in the reservoir (Figure 1). These direct measurements of diffusive fluxes were the first since the ones of Galy-Lacaux *et al.* [1997] in 1994 and 1995. In addition, in May 2003, diffusive fluxes were also measured in two natural sites for comparison: in a small confined march called Pri-Pri Yi-Yi and in the Kourou tidal river, which catchment is very similar to the one of the Sinnamary River. Fluxes were measured with three different floating chambers that were deployed simultaneously from a small boat that was left drifting during measurement to avoid creation of artificial turbulence. Two small chambers (volume: 16,5 L, surface: 0.2 m<sup>2</sup>), connected to gas analyzers for CO<sub>2</sub> and CH<sub>4</sub> were deployed during 5 min, at least on triplicate at each site, but most of the time five times. CO<sub>2</sub> was detected with a Non Dispersive Infra Red analyzer (CIRAS-2SC, PP system) and CH<sub>4</sub> with a Fourier Transformation Infra Red analyzer (GASMET DX-4010, Temet instruments). The gas ana-



**Table 1.** Carbon and Oxygen Concentrations Measured in the Petit Saut System Since Impounding<sup>a</sup>

	Petit Saut Reservoir, Station Roche Genipa						
	Epilimnion			Hypolimnion			Measurement Period
	Average	Range <sup>b</sup>	Number	Average	Range	Number	
O <sub>2</sub>	159	1–265	1521	0	0	–	Jan 1994 to Dec 2003
CH <sub>4</sub>	24	0.1–275	194	300	0.3–1300	483	Nov 1994 to Dec 2003
CO <sub>2</sub>	140	4–773	105	475	157–1516	175	Jan 1997 to Dec 2003 (except 2002)
TA	128	44–260	96	205	52–538	240	Jan 1999 to Dec 2002
POC	128	19–755	266	43	4–238	515	Jan 1994 to Dec 2002
DOC	430	242–603	166	475	178–987	414	June 1997 to Dec 2002

	Sinnamary River Downstream of the Dam						
	Immediately Downstream <sup>c</sup>			40 km Downstream, Station Pointe Combi			Measurement Period
	Average	Range	Number	Average	Range	Number	
O <sub>2</sub>	203	43–256	138	130	31–206	226	Feb 1995 to Dec 2002
CH <sub>4</sub>	73	8–371	190	37	0–293	185	Dec 1995 to Dec 2003
CO <sub>2</sub>	359	127–674	77	324	43–674	78	Dec 1996 to Dec 2003
TA	NM	NM	NM	139	66–290	60	Jan 1999 to Dec 2002
POC	50	33–81	12	107	35–314	66	July 1998 to Dec 2002
DOC	461	413–510	21	402	213–595	42	July 1998 to Dec 2002

<sup>a</sup>All values are given in  $\mu\text{mol L}^{-1}$ . In the reservoir, reference station is Roche Genipa; the epilimnion corresponds to samples at the surface and at 5 m depth and the hypolimnion to samples 5 m each from 10 to 35 m depths.

<sup>b</sup>In the epilimnion, the maximum carbon and minimum oxygen concentrations correspond to the first year after impounding.

<sup>c</sup>NM is not measured.

lyzers were calibrated with certified CO<sub>2</sub> and CH<sub>4</sub> gas standards at the beginning of each cruise. In addition, the GASMET cell was purged and zero was checked with nitrogen every day. Fluxes were calculated from the slope of the partial pressure of the gas in the chambers versus time. Flux data were accepted or rejected according to the procedure described by Lambert and Fréchet [2004]. The third chamber was larger (volume: 30 L, surface: 0.2 m<sup>2</sup>) and equipped with a rubber stopper that allowed gas sampling with a syringe and needle. This chamber was deployed during 30 min on the Sinnamary River and 60 min on the reservoir where fluxes are lower. Four 50 ml gas samples were taken at regular intervals (every 10 or 20 min) in the chamber and immediately transferred to N<sub>2</sub>-pre-flushed 10 ml serum vials. The CH<sub>4</sub> contents of the gas samples were then analyzed by means of GC-FID after correction for dilution factor in the gas sampling vials of 5/6. Fluxes were calculated from the linear regression of gas content versus time ( $R^2 > 0.85$ ,  $n = 4$ ).

#### 2.4. Estimation of Diffusive Gas Fluxes From Concentrations

[10] For most flux measurements, CO<sub>2</sub> and CH<sub>4</sub> concentrations in the surface water and in the atmosphere were also determined. This allowed the computation of the gas transfer velocity  $k$ , which was normalized to a Schmidt number of 600 [Wanninkhof, 1992]. In addition, for all the fluxes, wind speed was measured at 1 m height with a portable anemometer and was recalculated at 10 m height using the formulation of Amorcho and DeVries [1980]. In addition, in December 2003, rainfall was measured with a meteorological station near the dam. These data were used to establish the relationships between  $k_{600}$  wind speed and rainfall for the reservoir (F. Guérin et al., Gas transfer

velocities of CO<sub>2</sub> and CH<sub>4</sub> in a tropical reservoir and its river downstream, submitted to *Environmental Science and Technology*, 2005, hereinafter referred to as Guérin et al., submitted manuscript, 2005). These relationships were then combined to the average meteorological data recorded since January 2003 to calculate a value of  $k_{600}$  representative for the average conditions in Petit Saut. This  $k_{600}$  value was finally applied to the surface concentrations monitored monthly since impounding at station Roche Genipa in order to reconstruct the long-term CO<sub>2</sub> and CH<sub>4</sub> fluxes from the reservoir. Finally, the annual diffusive fluxes were calculated using a reservoir surface defined as a linear function of the maximum water level: 360 km<sup>2</sup> for a water level of 35 m and 260 km<sup>2</sup> for a water level of 30 m.

#### 2.5. Measurements of Bubbling Fluxes

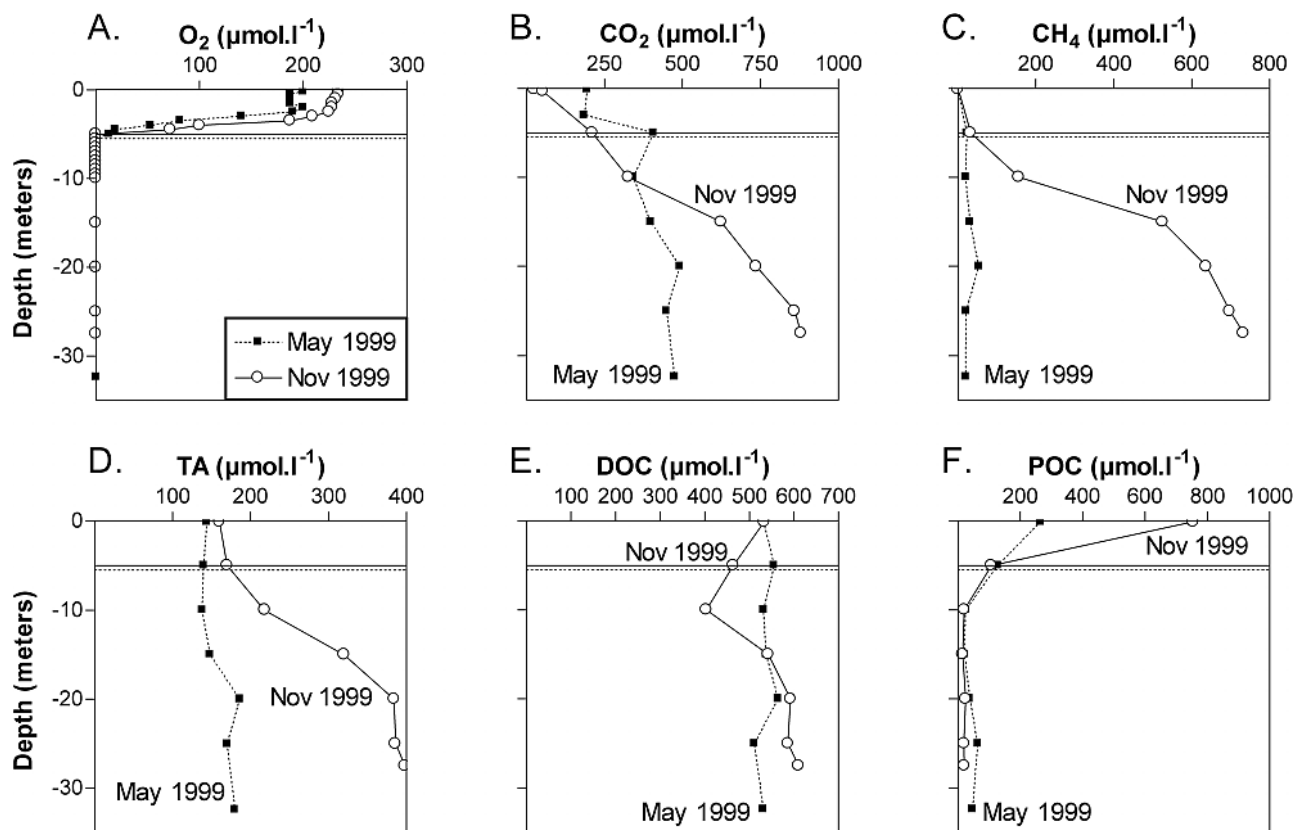
[11] In addition to the funnel measurement by Galy-Lacaux et al. [1999] in 1994 ( $n = 35$ ) and 1997 (134), bubbling fluxes were measured on 5 occasions in December 2003 at the same shallow stations. At a given depth of the water column, lines of 5 funnels of 0.7 m diameter each, were deployed during approximately 10 hours [Rosa et al., 2003]. The gas was collected in water-filled glass bottles connected to the funnels. The gas volume was recorded and the CH<sub>4</sub> and CO<sub>2</sub> concentrations were determined by GC analysis in the laboratory.

### 3. Results and Discussion

#### 3.1. Distributions of Oxygen and Carbon Species in the Reservoir

##### 3.1.1. Depth Profiles in the Reservoir

[12] Figure 2 shows classical distribution of oxygen and carbon species in the water column of the reservoir at

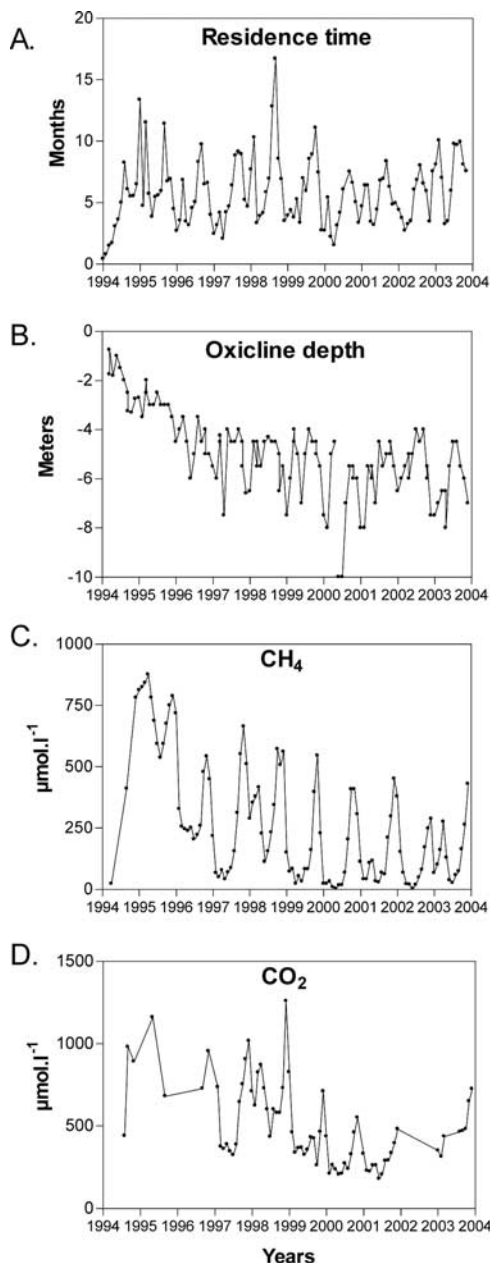


**Figure 2.** Examples of vertical profiles of (a) oxygen, (b) carbon dioxide, (c) methane, (d) total alkalinity (TA, mostly  $\text{HCO}_3^-$ ), (e) dissolved organic carbon (DOC), and (f) particulate organic carbon (POC) at the reference station Roche Genipa measured in May 1999 (high waters, solid squares) and November 1999 (low water, open circles).

station Roche Genipa during the dry and the wet seasons. The decomposition of organic matter in anoxic conditions at the reservoir bottom (flooded soil, leaves and sedimentary material) leads to high concentrations of CO<sub>2</sub> and CH<sub>4</sub> in the hypolimnion (Figures 2b and 2c). Concentrations are highest during the dry period (November), when residence time of water is longer. During the dry period, CO<sub>2</sub>/CH<sub>4</sub> ratios are close to one in the hypolimnion (Figures 2b and 2c), which reveals the predominance of methanogenesis. By contrast, during the wet season, CO<sub>2</sub> becomes much higher than CH<sub>4</sub>, owing to important inputs from the watershed. At the oxicleine, methane oxidation consumes a large part of the CH<sub>4</sub> diffusing upward [Dumestre et al., 1999], resulting to much lower concentrations at the water surface. This illustrates the major role played by methanotrophic bacteria in regulating the diffusive CH<sub>4</sub> flux from the reservoir surface [Richard et al., 2004]. Similarly, CO<sub>2</sub> is consumed by primary production in the epilimnion. Phytoplankton and phototrophic bacterioplankton (anaerobic green bacteria located just below the oxicleine and revealed by the presence of bacteriochlorophyll) are the main primary producers in Petit Saut [Vaquer et al., 1997; Dumestre et al., 1999]. Primary production is also reflected by the vertical distribution of POC in Figure 2f, which shows a maximum in the

epilimnion, particularly pronounced in November when CO<sub>2</sub> was minimum at the water surface (Figure 2b). The distribution of TA (Figure 2d) also shows an increase with depth during the dry season, which can be attributed to the occurrence of Iron reduction in the anoxic hypolimnion [Richard et al., 2000; Peretyazhko et al., 2005].

[13] DOC concentrations are naturally high in the Sinnamary River and Petit Saut reservoir, due to the contribution of humic substances leaching from forest soils [Richard et al., 2000]. This high-DOC background often masks some small changes due to internal biogeochemical processes in the reservoir. However, during the dry season, a significant trend can often be observed as illustrated by the November DOC profile in Figure 2e. A first DOC maximum occurs at the surface and is presumably related to the concomitant high-POC values and high primary production. Intense releases of DOC during phytoplankton blooms have been reported in the ocean [Meon and Kirchman, 2001] and in reservoirs [Mash et al., 2004]. A second DOC maximum is located in the anoxic hypolimnion and suggests a source at the reservoir bottom. Indeed, hydrolysis and fermentation during anaerobic decomposition of organic matter is known to produce DOC compounds that are not easily consumed by bacteria in anoxic conditions [Alperin et al., 1994;



**Figure 3.** Temporal changes in depth-integrated gas concentrations in the reservoir at station Roche Genipa since impounding in 1994: (a) average residence time of water in the reservoir, (b) depth of the oxicleine (in May 2003; the whole water column was oxygenated), (c) methane concentrations, and (d) carbon dioxide concentrations.

*Kristensen et al.*, 1995]. Through this process, the transport of DOC from the anoxic hypolimnion could fuel aerobic respiration and photo-oxidation in the epilimnion and in the estuary.

### 3.1.2. Downstream Concentrations

[14] Because of the presence of the submerged wall, the water column gets mixed before passing through the turbines. The average oxygen concentration is 159  $\mu\text{mol L}^{-1}$

in the epilimnion (Table 1) and 30  $\mu\text{mol L}^{-1}$  in the turbined water [Gosse *et al.*, 2005]. This corresponds to a contribution of 18% of the epilimnion and 82% of the hypolimnion to the turbined water. This mixing ratio is consistent with the relative contribution of the epilimnion (6–7 m) to the whole water column at the reservoir maximum depth (30–35 m). When meeting more oxygen at the dam outlet (aerating weir and bottom gates), drastic changes of the chemical composition of the water occur because of two major processes: gas exchange and oxidation of reduced compounds. This can be seen in Table 1, where concentrations in the Petit Saut reservoir are compared with those in the Sinnamary river 0.8 km and 40 km downstream of the dam (Station Pointe Combi). When passing over the weir, waters lose a large fraction of their CO<sub>2</sub> and CH<sub>4</sub> and get reoxygenated. Fe<sup>2+</sup> concentrations also rapidly decrease to nearly zero at the station Pointe Combi [Richard *et al.*, 2000]. This Fe oxidation, is accompanied by a decrease in TA from the reservoir to station Pointe Combi (Table 1).

### 3.1.3. Long-Term Change in Average Water Column Concentrations in the Reservoir

[15] Figure 3 shows how depth-integrated gas concentrations in the reservoir and the oxicleine depth have varied at station Roche Genipa since impounding. The first trend is a net decrease in CO<sub>2</sub> and CH<sub>4</sub> concentrations and in the oxicleine depth during the first 3 to 4 years after impounding (until 1997–1998). This trend had been predicted by Galy-Lacaux *et al.* [1999] and Delmas *et al.* [2001] when they extrapolated measurements during the first 2 years to respectively 20 and 100 years. After the year 1998, gas concentrations and the oxicleine depth stabilized or decreased much more slowly. Gas concentrations followed a seasonal cycle well correlated with the water residence time, which confirms that the major gas source is located at the reservoir bottom (flooded soil, leaves and sedimentary material). The year 2000 was particularly wet and the whole reservoir water body became exceptionally oxygenated with very low CH<sub>4</sub> concentrations during 3 months. Such mixing events are however uncommon and the oxicleine generally remains in place even during the wet seasons.

## 3.2. CO<sub>2</sub> and CH<sub>4</sub> Atmospheric Fluxes

[16] As described in earlier studies [Galy-Lacaux *et al.*, 1997, 1999] gaseous emissions from the Petit Saut system occur through four different pathways: (1) the diffusive flux from the reservoir surface; (2) the bubbling flux, which is restricted to the shallowest (<10 m depth) areas of the reservoir; (3) the degassing at the aerating weir immediately downstream of the dam; and (4) the diffusive flux from the Sinnamary River downstream of the dam.

### 3.2.1. Diffusive Fluxes From the Reservoir

#### 3.2.1.1. Diffusive Fluxes Measured

##### With Floating Chambers

[17] Diffusive fluxes measured with the floating chambers in May and December 2003 ranged between  $14 \pm 3$  and  $131 \pm 116 \text{ mmol m}^{-2} \text{ d}^{-1}$  for CO<sub>2</sub> and between  $2.7 \pm 1.6$  and  $7.7 \pm 8.8 \text{ mmol m}^{-2} \text{ d}^{-1}$  for CH<sub>4</sub> (Table 2). Fluxes were significantly lower in flooded forest sites than in open waters. This was not due to lower concentrations in the flooded forest waters but rather to lower wind speeds and

**Table 2.** Diffusive Fluxes Measured With Floating Chambers in the Petit Saut Reservoir and in the Sinnamary River Downstream in May and December 2003

	CO <sub>2</sub>			CH <sub>4</sub>		
	Flux, mmol m <sup>-2</sup> d <sup>-1</sup>	SD	n	Flux, mmol m <sup>-2</sup> d <sup>-1</sup>	SD	n
<i>Reservoir (Surface 300 km<sup>2</sup>)</i>						
Flooded forest						
May 2003	57	28	16	3.1	2.3	12
Dec 2003	14	3	4	–	–	–
Open waters						
May 2003	133	116	67	7.7	8.8	32
Dec 2003	131	110	98	2.7	1.6	22
<i>Sinnamary River</i>						
Upstream of the reservoir						
May 2003	461	288	3	0.48	–	1
Dec 2003	30	–	1	–	–	–
<40 km downstream of the reservoir (surface 4 km <sup>2</sup> )						
May 2003	1003	574	21	59.3	25.3	12
Dec 2003	928	370	20	123.3	60.8	16
>40 km downstream of the reservoir (surface 17 km <sup>2</sup> )						
May 2003	704	436	14	0.6	0.4	9
Dec 2003	750	317	13	2.6	1.0	5

lower gas transfer velocities during measurements. Wind speed and rainfall are highly variable daily at the Petit Saut and much more windy and rainy events had been sampled in open waters than in flooded forest. When comparing these chamber measurements with those of *Galy-Lacaux et al.* [1997], the CO<sub>2</sub> fluxes were the same order of magnitude in 2003 and in 1994–1995 ( $125 \pm 83$  mmol m<sup>-1</sup> d<sup>-1</sup>). By contrast, CH<sub>4</sub> diffusive fluxes have decrease by one order of magnitude compared to the 1994–1995 period ( $51 \pm 78$  mmol m<sup>-2</sup> d<sup>-1</sup>).

### 3.2.1.2. Diffusive Fluxes Calculated From Concentrations

[18] On the reservoir, the  $k_{600}$  dependence to wind speed matched well the formulation of *Cole and Caraco* [1998] for lakes (Guérin et al., submitted manuscript, 2005). An additional contribution of rainfall to  $k_{600}$ , which matched the formulation of *Ho et al.* [1997], could also be calculated. The  $k_{600}$  computed with one year meteorological average data was 3.3 cm h<sup>-1</sup>, for a wind speed of 1.7 m s<sup>-1</sup> and a

rainfall of 0.4 mm h<sup>-1</sup>. In Table 3, we show how representative is the station Roche Genipa for the whole reservoir and we compare measured and calculated fluxes. During the chamber flux measurements, the CO<sub>2</sub> and CH<sub>4</sub> surface concentrations were slightly lower at station Roche Genipa than at all stations in the reservoir. This discrepancy was due to much higher concentration (CO<sub>2</sub> up to 198 μmol L<sup>-1</sup> and CH<sub>4</sub> up to 21 μmol L<sup>-1</sup>) in only two flooded forest sites. When comparing chamber fluxes at Roche Genipa and at all stations on the reservoir, there was no significant ( $p > 0.05$ ) difference for CO<sub>2</sub>. By contrast, CH<sub>4</sub> fluxes were significantly lower ( $p < 0.001$ ) at Roche Genipa by a factor 1.8. This discrepancy could partly be explained by the lower wind speed and lower  $k_{600}$  during measurements at Roche Genipa. Station Roche Genipa is thus well representative of the reservoir average for CO<sub>2</sub> flux but tend to underestimate the CH<sub>4</sub> flux by about 50%, according to Table 3 and after a normalization to the same  $k_{600}$ . At station Roche Genipa, for the year 2003, calculated fluxes

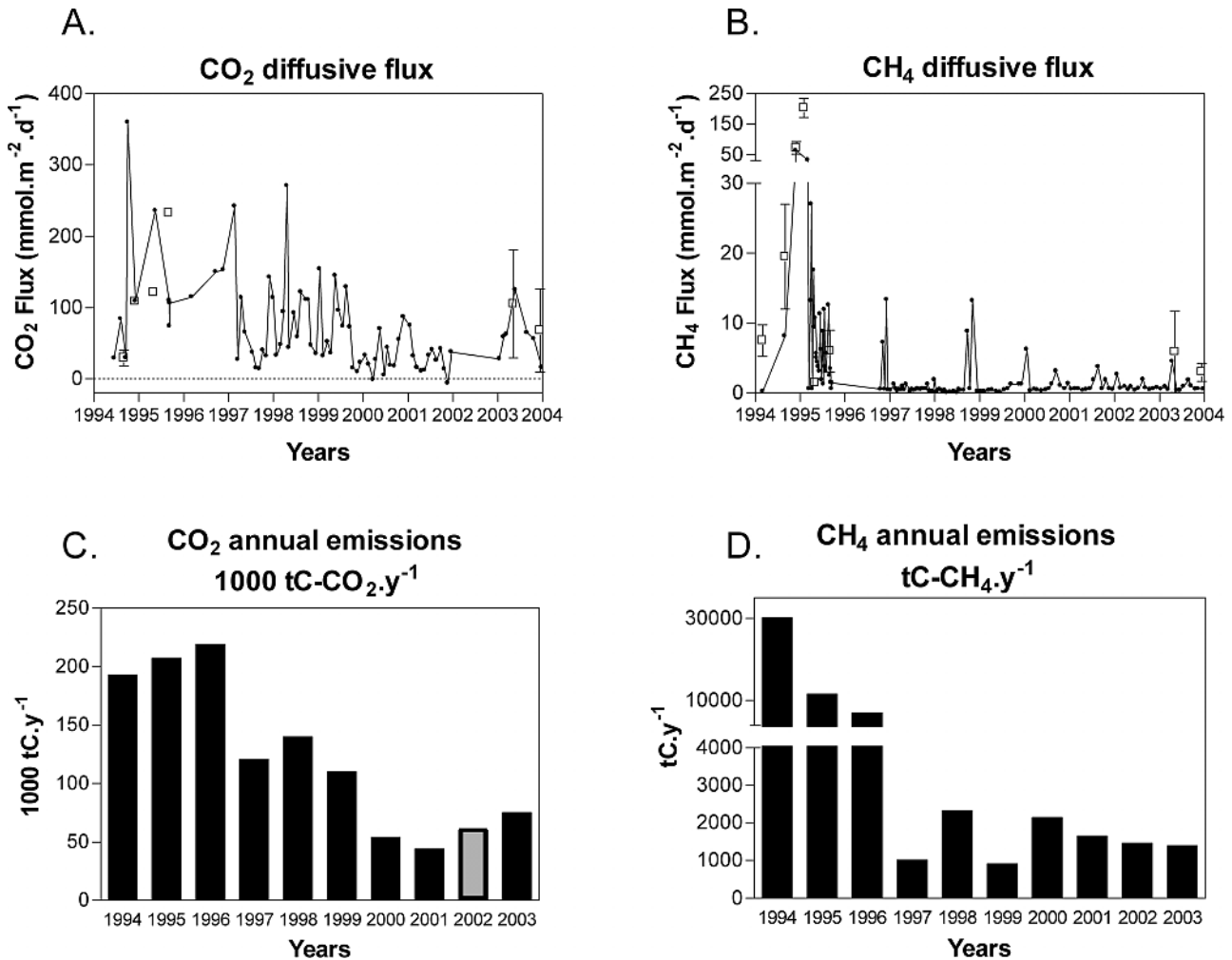
**Table 3.** Comparison of CO<sub>2</sub> and CH<sub>4</sub> Fluxes in the Petit Saut Reservoir Measured With Chambers at All Stations and at Station Roche Genipa During the Two 2003 Surveys and Calculated Using the Average Wind Speed and Rainfall and the Surface Concentration at Station Roche Genipa

	Direct Flux Measurements in 2003		Calculated Fluxes for 2003 at Station Roche Genipa	
	All Stations	Station Roche Genipa	Parameters Used for Calculation	Calculated Fluxes
CO <sub>2</sub> concentrations, μmol L <sup>-1</sup>	108 ± 63 (33)	73 ± 28 (4)	70 ± 43 (12)	
CO <sub>2</sub> fluxes, mmol m <sup>-2</sup> d <sup>-1</sup>	108 ± 77 (175)	75 ± 36 (24)		47 ± 25 (12)
CH <sub>4</sub> concentrations, μmol L <sup>-1</sup>	2.5 ± 2.6 (56)	1.7 ± 0.8 (2)	1.1 ± 0.8 (14)	
CH <sub>4</sub> fluxes, mmol m <sup>-2</sup> d <sup>-1</sup>	2.9 ± 1.8 (58)	1.7 ± 0.7 (8)		1.2 ± 0.7 (14)
Wind speed at 10 m, m s <sup>-1</sup>	2.4 ± 1.7 (32)	2.1 ± 1.4 (8)	1.7 <sup>a</sup>	
$K_{600}$ , cm h <sup>-1</sup>	4.6 ± 3.2 (65)	4.3 ± 2.7 (6)	3.3 <sup>b</sup>	

<sup>a</sup>Average wind speed measured continuously during the year 2003 by a meteorological station.

<sup>b</sup>Average gas transfer velocity obtained with the average wind speed and rainfall and a parameterization based on all flux measurements (Guérin et al., submitted manuscript, 2005).





**Figure 4.** Temporal change in diffusive fluxes since impounding in 1994: (a) CO<sub>2</sub> diffusive fluxes, (b) CH<sub>4</sub> diffusive fluxes measured in the whole reservoir (open squares; the error bars are the standard deviation due to spatial variations) and calculated at station Roche Genipa (solid circles; see text for calculation procedure), and resulting annual (c) CO<sub>2</sub> and (d) CH<sub>4</sub> emissions by diffusion from the reservoir surface. The gray bar is interpolated because no data were available.

were always lower than measured fluxes but this was due to wind speeds higher than the average during chambers measurements.

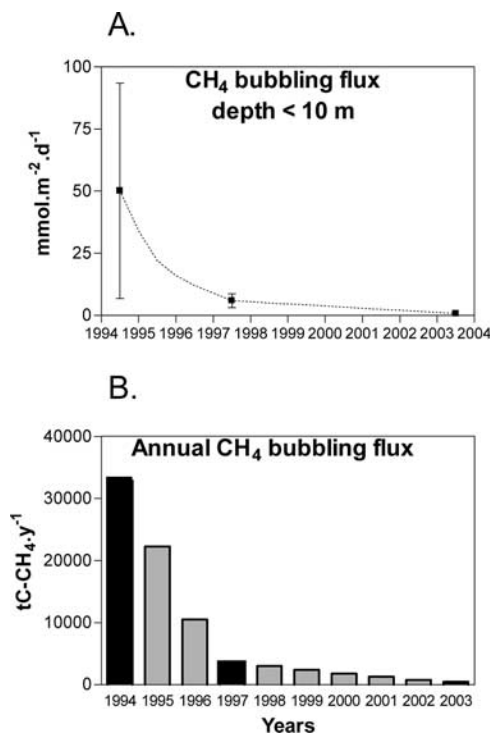
### 3.2.1.3. Long-Term Changes in Diffusive Fluxes

[19] CO<sub>2</sub> diffusive fluxes measured directly and calculated from surface concentrations varied between  $-10$  and  $+370$  mmol m<sup>-2</sup> d<sup>-1</sup> (Figure 4a). Undersaturation in CO<sub>2</sub> with respect to the atmosphere was observed occasionally in February 2000 and December 2002. This long-term decrease results from changes in the balance between the decomposition of the flooded biomass at the reservoir bottom and CO<sub>2</sub> uptake by phytoplankton in the epilimnion. Since Chlorophyll-*a* concentrations were maximal in 1994 and 1995 and then decreased with time [Vaquer *et al.*, 1997], a decrease of CO<sub>2</sub> diffusion from the reservoir hypolimnion must explain most of the decreasing trend in CO<sub>2</sub> diffusive fluxes. This is also confirmed by the long-term decrease in average CO<sub>2</sub> concentration in the

reservoir water column (Figure 3d). However, 10 years after impounding, the reservoir is still not a net remover of CO<sub>2</sub> from the atmosphere. The integrated annual CO<sub>2</sub> diffusive emissions (Figure 4c) decreased from  $\sim 200 \times 10^3$  t yr<sup>-1</sup> C-CO<sub>2</sub> in 1994–1996 to less than  $70 \times 10^3$  t yr<sup>-1</sup> C-CO<sub>2</sub> in 2000. Since the year 2000, CO<sub>2</sub> emissions appear to have stabilized with an interannual variation linked to hydrology and changes in residence time.

[20] CH<sub>4</sub> diffusive fluxes (Figure 4b) using combined measured and calculated data were maximum in February 1995, reaching  $200$  mmol m<sup>-2</sup> d<sup>-1</sup> [Galy-Lacaux *et al.*, 1997]. They then decreased very suddenly to less than  $10$  mmol m<sup>-2</sup> d<sup>-1</sup> in May 1995, owing to the installation of a very active population of methanotrophic bacteria [Dumestre *et al.*, 1999]. After 1996, calculated CH<sub>4</sub> diffusive fluxes were most of the time lower than  $2$  mmol m<sup>-2</sup> d<sup>-1</sup>, corresponding to a methane concentration in the water around  $1$  μmol L<sup>-1</sup>. Higher diffusive fluxes





**Figure 5.** (a) Temporal change in CH<sub>4</sub> bubbling fluxes measured in 1994, 1997, and 2003 in the shallow part of the reservoir (depth <10 m); the line represents an exponential decay that was used for interpolation. (b) Resulting annual CH<sub>4</sub> bubbling emissions. Gray bars are interpolated data. Note that the CO<sub>2</sub> bubbling flux was negligible.

occurred occasionally during short events with higher surface concentrations in particular at the end of the dry season (November–December). At that time, the hypolimnion was rich in CH<sub>4</sub> and destratification of the water column occurred because of the first events of heavy rain and increasing river discharge and water current. These events of higher CH<sub>4</sub> concentrations in surface waters played a major role in the annual budget and decreased in frequency and magnitude with reservoir aging (Figure 4b). On the basis of combined data, yearly integrated diffusive fluxes (Figure 4d) decreased from ~30,000 t yr<sup>-1</sup> C-CH<sub>4</sub> in 1994 to ~7000 t yr<sup>-1</sup> C-CH<sub>4</sub> in 1996. Since 1997, diffusive CH<sub>4</sub> fluxes have stabilized at around 1 500 t yr<sup>-1</sup> C-CH<sub>4</sub>.

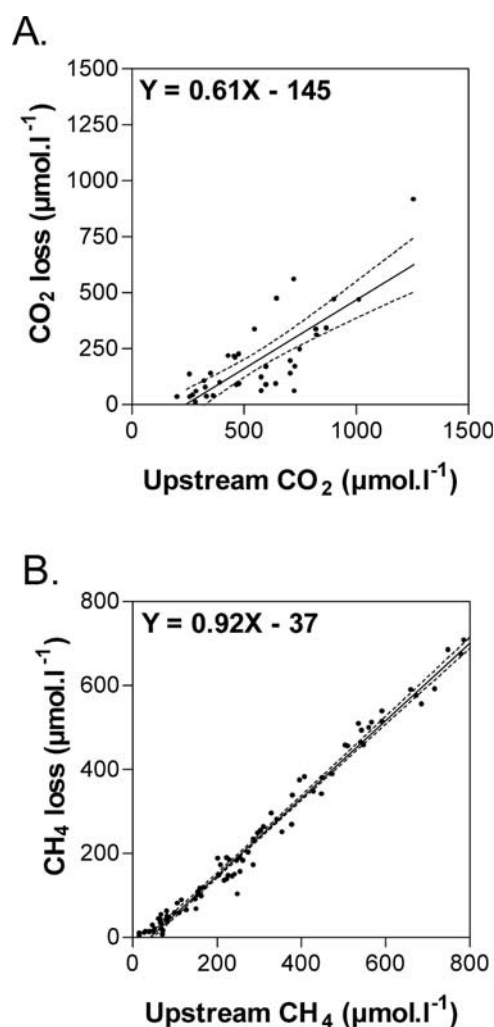
### 3.2.2. Bubbling Fluxes From the Reservoir

[21] Only few funnel measurements of bubbling fluxes have been performed at Petit Saut in 1994 ( $n = 35$ ), in 1997 ( $n = 134$ ) by *Galy-Lacaux et al.* [1999] and more recently in 2003 during our December survey ( $n = 5$ ). *Galy-Lacaux et al.* [1999] found bubbling to occur only at depths lower than 10 m. Gas bubbles contained 50 to 80% CH<sub>4</sub> with few amounts (<1%) of CO<sub>2</sub>. These authors reported a decrease of one order of magnitude of the average CH<sub>4</sub> bubbling flux from ~50 mmol m<sup>-2</sup> d<sup>-1</sup> in 1994 to ~6 mmol m<sup>-2</sup> d<sup>-1</sup> in 1997 (Figure 5a). Although only 5 funnel measurements were performed in 2003, fluxes at depth below 10 m were  $0.7 \pm 0.5$  mmol m<sup>-2</sup> d<sup>-1</sup>, that is, again one more order of

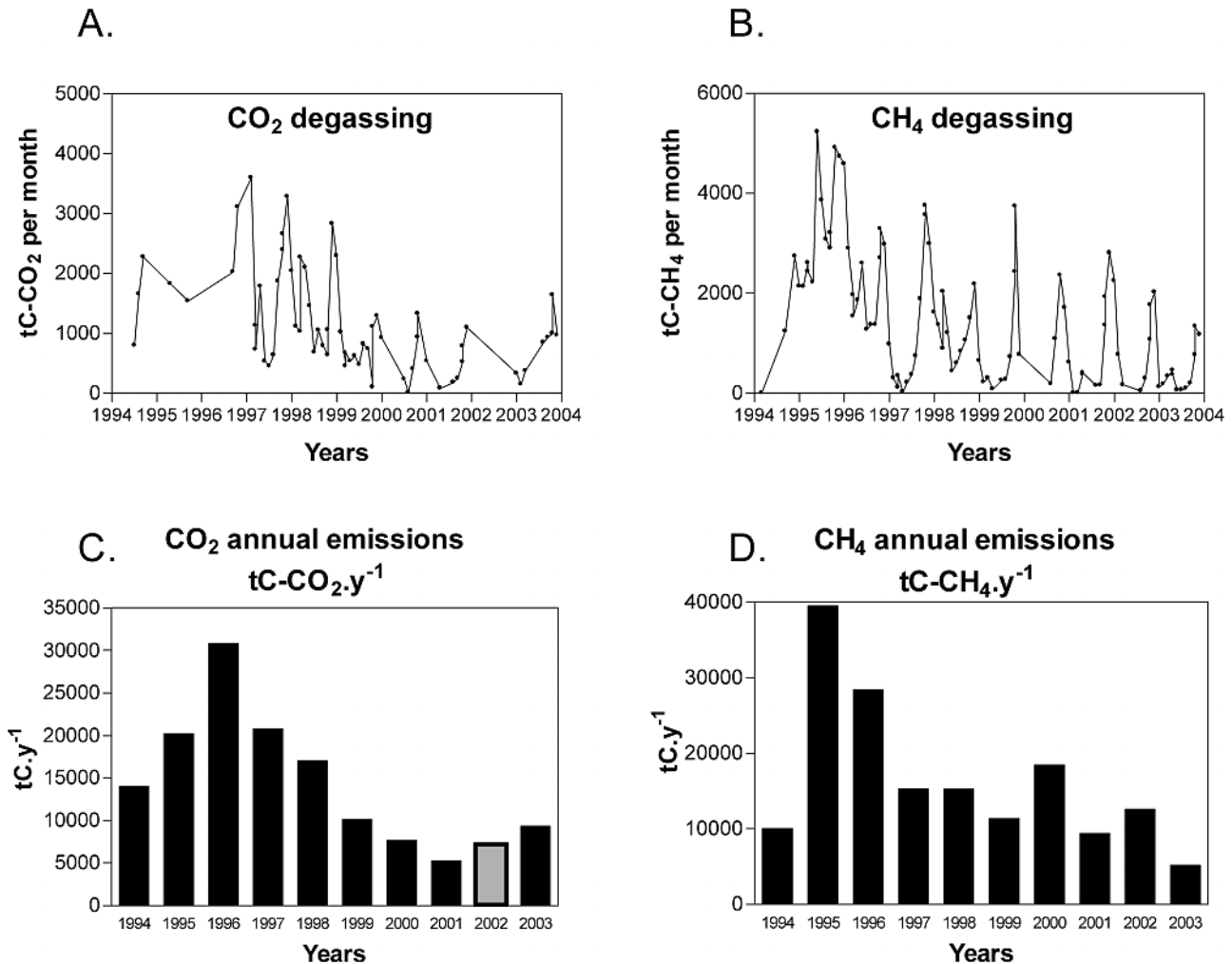
magnitude lower than in 1997. Consequently, the potential error in this emission term in 2003 is moderate compared to other terms. Fluxes were integrated using a reservoir surface with depths <10 m of 150 km<sup>2</sup> [*Galy-Lacaux et al.*, 1999]. Annual CH<sub>4</sub> emissions by bubbling (Figure 5b) show an exponential decrease from 32,000 t yr<sup>-1</sup> C-CH<sub>4</sub> in 1994 to ~440 t yr<sup>-1</sup> C-CH<sub>4</sub> in 2003. Although the CH<sub>4</sub> bubbling flux was similar to the diffusive flux in 1994, it has become a very minor component to the total flux 10 years after impounding. The CO<sub>2</sub> bubbling flux was negligible (<1%) compared to the diffusive flux, even the first year after impounding.

### 3.2.3. Degassing at the Outlet of the Dam

[22] In Petit Saut, there are two pathways of degassing at the outlet of the dam [*Gosse et al.*, 2000]: one continuous at the aerating weir that receives the turbined water and another one occurring only during the wet season and flood events at the jet and hydraulic jump downstream of the



**Figure 6.** Degassing at the aerating weir. Relationships between the (a) CO<sub>2</sub> and (b) CH<sub>4</sub> loss at the weir (depth-integrated concentrations in the reservoir (Roche Genipa) minus concentrations downstream of the weir approximately 2 weeks later) and the Roche Genipa concentration.

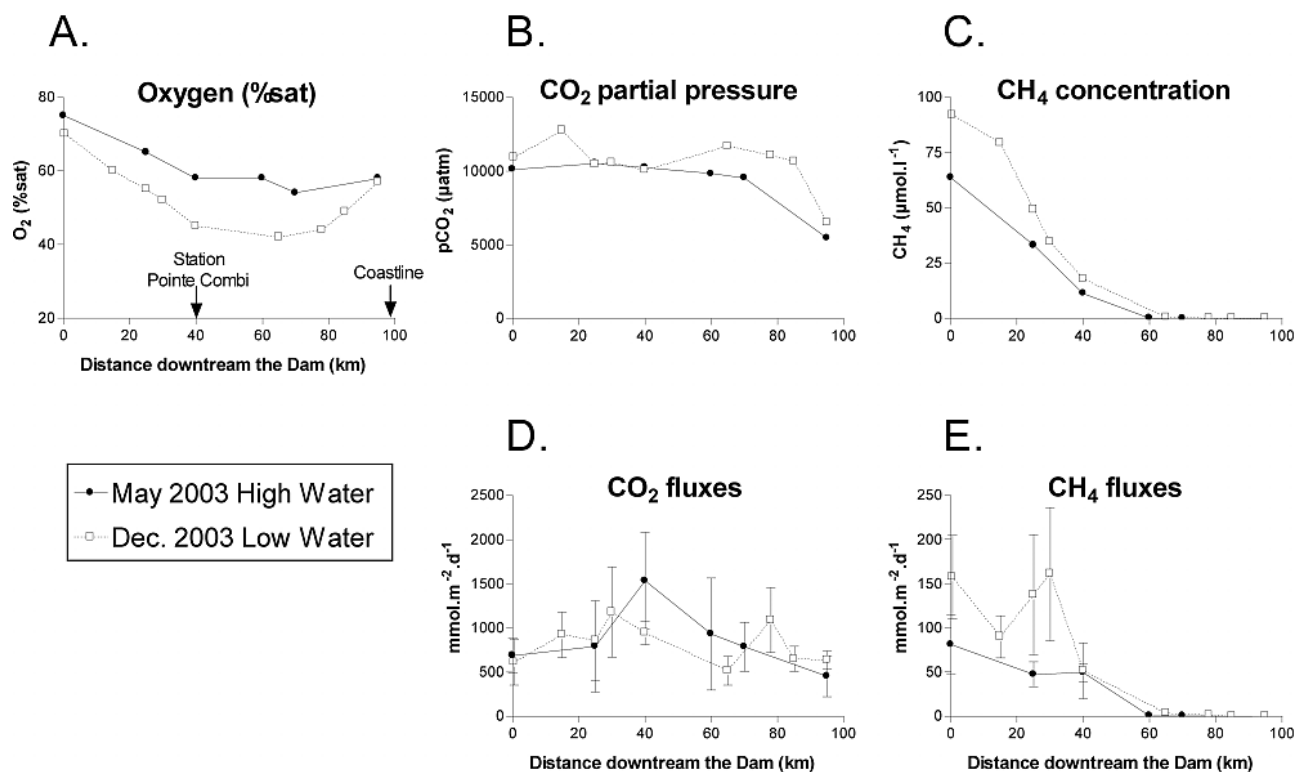


**Figure 7.** Temporal changes in the CO<sub>2</sub> and CH<sub>4</sub> degassing flux at the weir: (a and b) monthly fluxes and (c and d) annual fluxes.

bottom gates, when opened. In order to estimate the sum of these two degassing fluxes, we have compared the depth-integrated CO<sub>2</sub> and CH<sub>4</sub> concentrations at station Roche Genipa with those measured approximately 2 weeks later 0.8 km downstream of the dam. The number of concomitant data upstream and downstream was 78 for CH<sub>4</sub> (covering the whole 1995–2003 period) and 38 for CO<sub>2</sub> (from 1997 to 2003 with no data in 2002). We obtained linear correlations between the loss of CO<sub>2</sub> and CH<sub>4</sub> at the outlet of the dam (difference upstream-downstream) and the concentration upstream (Figure 6). These relationships reflect the efficiency of the weir for degassing. For CO<sub>2</sub> and CH<sub>4</sub> respectively, slopes were 0.6 and 0.9 and threshold concentrations were 250 and 45  $\mu\text{mol L}^{-1}$ . These results are consistent with those of Gosse *et al.* [1997] and Richard *et al.* [2004] who compared gas concentrations in the turbine and downstream of the weir and obtained degassing rates of 80% for CH<sub>4</sub> and 65% for CO<sub>2</sub>. Because of a partial lowering of the weir, this fraction has decreased to 65% for CH<sub>4</sub> since March 2003 [Delmas *et al.*, 2004; Gosse *et al.*, 2005]. The better degassing efficiency for CH<sub>4</sub> can be explained by its lower

solubility: at a temperature of 28 degrees, the solubility of CH<sub>4</sub> is  $\sim 25$  times lower than the one of CO<sub>2</sub>; thus, in the turbulent conditions of the weir, CH<sub>4</sub> escapes the water much more rapidly and in higher proportion than CO<sub>2</sub>.

[23] We have used the two relationships in Figure 6 together with the depth-integrated gas concentrations at station Roche Genipa (Figure 3) and the total discharge (turbines plus bottom gates, when opened) in order to calculate the CO<sub>2</sub> and CH<sub>4</sub> degassing fluxes since impounding (Figure 7). Similarly to other emissions terms, the degassing flux increased the first 2–3 years and then decreased. The annual CO<sub>2</sub> degassing (Figure 7c) was maximum in 1996 at  $30 \times 10^3 \text{ t yr}^{-1} \text{ C-CO}_2$ , when concentrations and turbined discharge also reached their maximum. Since 1999, CO<sub>2</sub> degassing has stabilized to less than  $10 \times 10^3 \text{ t yr}^{-1} \text{ C-CO}_2$  with some interannual variations due to hydrological conditions. Annual CH<sub>4</sub> degassing (Figure 7d) was maximum in 1995 ( $\sim 40 \times 10^3 \text{ t yr}^{-1} \text{ C-CH}_4$ ) when concentrations in the reservoir where maximum and then decreased to about  $10 \times 10^3 \text{ t yr}^{-1} \text{ C-CH}_4$  since 1997. These estimations are consistent at 10%



**Figure 8.** Longitudinal profiles of (a) O<sub>2</sub>, (b) partial pressure of CO<sub>2</sub>, (c) CH<sub>4</sub> concentration, (d) CO<sub>2</sub> fluxes, and (e) CH<sub>4</sub> fluxes measured in the Sinnamary River and estuary in May (solid circles) and December (open squares) 2003.

with those of *Richard et al.* [2004] who used the gas concentrations in the turbines rather than at station Roche Genipa.

### 3.2.4. Fluxes From the Sinnamary Tidal River and Estuary Downstream of the Weir

[24] CH<sub>4</sub> fluxes on the Sinnamary tidal river and estuary measured in 2003 were on average 90 mmol m<sup>-2</sup> d<sup>-1</sup> along the first 40 km downstream of the reservoir, and fell down to 0.6–2.6 mmol m<sup>-2</sup> d<sup>-1</sup> from 40 km to the ocean (Table 2 and Figure 8e). In a recent compilation of data in estuaries worldwide, *Abril and Borges* [2004] reported CH<sub>4</sub> water-air fluxes in the main channels of estuaries ranging between 0.007 and 0.41 mmol m<sup>-2</sup> d<sup>-1</sup>. The CH<sub>4</sub> fluxes measured in May 2003 in the two natural sites in French Guiana were within this range: 0.43 ± 0.25 mmol m<sup>-2</sup> d<sup>-1</sup> in the Pri-Pri Yi-Yi marsh and 0.37 ± 0.17 mmol m<sup>-2</sup> d<sup>-1</sup> in the Kourou tidal River. Also, the Sinnamary river section from 40 km downstream of the dam to the ocean emits CH<sub>4</sub> at rates that can be considered as natural for estuaries. This is however not the case for the first 40 km section of the Sinnamary River downstream of the dam, where the average CH<sub>4</sub> flux was ~200 times higher than the natural ones. The CH<sub>4</sub> in this section of the river originates from the reservoir and corresponds to the threshold concentration in Figure 6a that has not degassed at the aerating weir. In the river, this CH<sub>4</sub> is partly oxidized and partly emitted to the atmosphere with an emission/oxidation ratio ranging between 0.35 and 0.62 [*Galy-Lacaux et al.*, 1997; *Gosse et al.*, 2000, 2005; *Richard et al.*, 2004]. With a surface of 4 km<sup>2</sup>, this river

section emitted 2.7 and 5.9 t d<sup>-1</sup> C-CH<sub>4</sub> in May and December 2003 respectively, which corresponds to an average annual emission of 1560 t yr<sup>-1</sup> C-CH<sub>4</sub>. This additional CH<sub>4</sub> source in the Sinnamary estuary was similar to the diffusive flux from the reservoir surface in 2003 (1390 t yr<sup>-1</sup> C-CH<sub>4</sub>; Figure 4d). In order to calculate the annual emissions since impounding, we used the monthly concentrations monitored downstream of the weir (Table 1), the water discharge and an average emission/oxidation ratio of 0.5 [*Gosse et al.*, 2000, 2005]. Except in 1994, CH<sub>4</sub> emissions from the Sinnamary river downstream of the dam were around 1500 t yr<sup>-1</sup>, close to the 2003 value (Table 4).

[25] The CO<sub>2</sub> fluxes measured in May and December 2003 in the Sinnamary tidal river and estuary (Figure 8d) were the first since impounding. They were on average for the two cruises 950 mmol m<sup>-2</sup> d<sup>-1</sup> in the first 40 km downstream of the dam and 720 mmol m<sup>-2</sup> d<sup>-1</sup> downstream of 40 km. pCO<sub>2</sub> were close or higher than 10,000 µatm (Figure 8b), except at the station located at the limit of the coast where it was around 6000 µatm (salinity at this point was around 3 for both surveys). Like for CH<sub>4</sub>, these CO<sub>2</sub> fluxes are higher than those from the literature for estuaries, where fluxes converge to about 100 mmol m<sup>-2</sup> d<sup>-1</sup> [*Abril and Borges*, 2004]. The highest pCO<sub>2</sub> reported so far in upstream regions of estuaries was 7800 µatm in the Altamaha (Georgia, United States), an estuary greatly influenced by inputs from tidal marshes [*Cai et al.*, 1999] and 9500 µatm in the Scheldt (Belgium), an estuary that receives large amounts of untreated sewage [*Frankignoulle et al.*,

**Table 4.** Synthesis of Computed CO<sub>2</sub> and CH<sub>4</sub> Emissions in Petit Saut From 1994 to 2003<sup>a</sup>

Year	CO <sub>2</sub> Emissions			CH <sub>4</sub> Emissions			
	Diffusive <sup>b</sup>	Degassing <sup>c</sup>	Estuary <sup>d</sup>	Diffusive <sup>b</sup>	Bubbling <sup>c</sup>	Degassing <sup>c</sup>	Estuary <sup>d</sup>
1994	193,000	14,000	75,000	30,150	32,850	10,000	190
1995	207,000	20,200	75,000	11,620	22,240	39,440	1,850
1996	219,000	30,800	75,000	6,990	10,470	28,400	2,800
1997	121,000	20,800	75,000	1,020	3,470	15,280	2,340
1998	140,000	17,100	62,500	2,330	2,980	15,230	1,240
1999	110,000	10,100	50,000	910	2,370	11,320	1,170
2000	54,000	7,700	37,500	2,150	1,770	18,460	1,350
2001	44,000	5,300	37,500	1,660	1,260	9,380	890
2002	60,000	7,300	37,500	1,470	750	12,570	1,540
2003	75,000	9,300	37,500	1,390	440	5,180	1,560

<sup>a</sup>Emissions in t yr<sup>-1</sup> C.

<sup>b</sup>Diffusive flux from the reservoir measured using floating chambers and calculated from surface concentrations at station Roche Genipa.

<sup>c</sup>Degassing flux at the reservoir outlet (aerating weir and bottom gates) calculated as the difference in gas concentrations in the reservoir (average water column at Roche Genipa) and downstream of the aerating weir.

<sup>d</sup>Diffusive flux from the Sinnamary tidal river and estuary downstream of the weir; for CH<sub>4</sub>, the totality of the flux in the 0–40 km river section was considered, and annual fluxes were calculated using the monthly concentrations downstream of the weir, the water discharge, and an emission/oxidation ratio of 0.5. For CO<sub>2</sub>, only half of the 2003 flux from the whole estuary (0 km to ocean) was considered, and annual fluxes were extrapolated to previous years on the basis of the long-term changes in concentrations at station Pointe Combi (40 km downstream of the dam).

<sup>e</sup>Bubbling flux from the shallow (<10 m) area of the reservoir interpolated from funnel measurements in 1994, 1997, and 2003. The CO<sub>2</sub> bubbling flux was always negligible (<1% of the total).

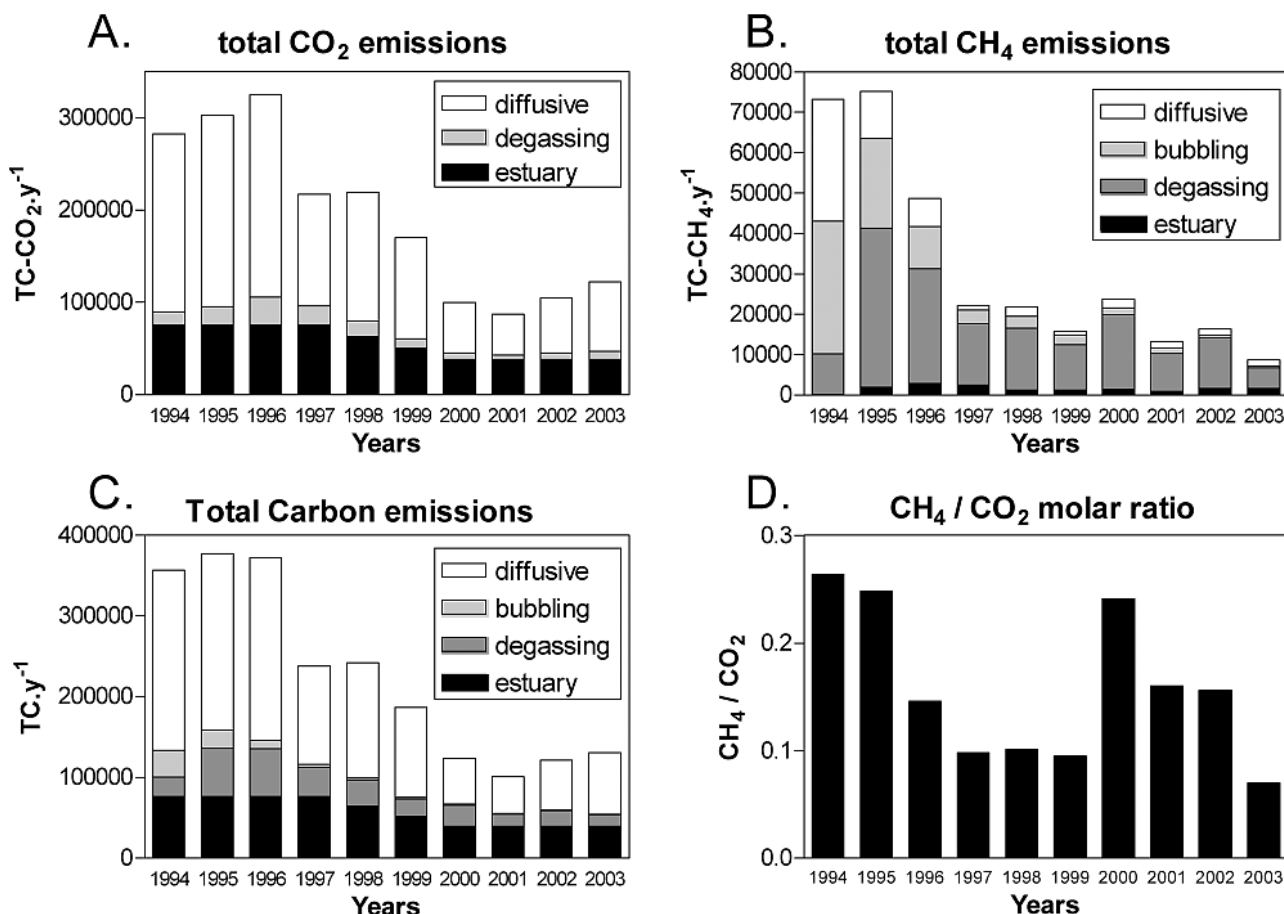
1998; *Abril et al.*, 2000]. In May 2003, we measured CO<sub>2</sub> fluxes of  $436 \pm 55$  mmol m<sup>-2</sup> d<sup>-1</sup> in the Pri-Pri Yi-Yi marsh and  $370 \pm 32$  mmol m<sup>-2</sup> d<sup>-1</sup> in the Kourou tidal River. This last value is probably in the upper range for the Kourou tidal river since it was measured under a storm and heavy rain. Nevertheless, the CO<sub>2</sub> atmospheric fluxes in the Sinnamary River estuary were in 2003 at least 2 times the ones of similar natural systems. Using data from Table 2 we calculated a CO<sub>2</sub> emission of 200 t d<sup>-1</sup> for each cruise, which corresponds to an annual average of 75,000 t yr<sup>-1</sup> C-CO<sub>2</sub>, that is, the same value as the diffusive CO<sub>2</sub> flux from the reservoir surface (Figure 4c). The input of dissolved CO<sub>2</sub> from the reservoir, as calculated from concentrations downstream of the weir and water discharge, contributed to only 25% and 21% of these emissions in May and December 2003 respectively. In order to maintain high pCO<sub>2</sub> and CO<sub>2</sub> fluxes over such a long distance in the estuary, additional sources must occur. Like most estuaries [*Gattuso et al.*, 1998], the Sinnamary tidal river and estuary must also be the site of intense heterotrophic activity, partly fueled by input of labile organic matter from the reservoir: either DOC leaking from the flooded soil and biomass or autochthonous POC and DOC produced in the reservoir (see section 3.1.1). A detailed budget of organic carbon in the Sinnamary estuary is however difficult to establish because of the number of unconstrained processes occurring in this area including flocculation and deflocculation, sedimentation and resuspension, etc. . . Finally lateral natural inputs of CO<sub>2</sub> from mangrove at the lower reach of the Sinnamary estuary probably also significantly contribute to the budget. Mangrove surrounding waters often show pCO<sub>2</sub> higher than 6000 μatm [*Borges et al.*, 2003; *Bouillon et al.*, 2003]. On the basis of all these considerations, we attribute one half of the total CO<sub>2</sub> fluxes in the Sinnamary estuary in 2003 to the presence of the Petit Saut reservoir upstream and the other half to natural processes. Although the natural CO<sub>2</sub> flux from the estuary can be considered as constant, the contribution of the reservoir has probably decreased with time since

impounding. The long-term CO<sub>2</sub> emission from the estuary can be extrapolated to previous years on the basis of the monitoring at station Pointe Combi (40 km downstream), which, for CO<sub>2</sub>, covers the 1997–2003 period (Table 1). Annual average CO<sub>2</sub> concentrations decreased linearly from 555 μmol L<sup>-1</sup> in 1997 to a stable value of  $274 \pm 56$  μmol L<sup>-1</sup> since the year 2000 (not shown). We reconstructed the CO<sub>2</sub> annual fluxes from the estuary due to the reservoir by assuming they were proportional to the annual average CO<sub>2</sub> concentrations (Table 4). For the years 1994–1996 when no concentration were available, we assumed this CO<sub>2</sub> fluxes were the same as in 1997.

### 3.2.5. Total Emissions and Relative Contribution of Each Term

[26] Figure 9 and Table 4 summarize the CO<sub>2</sub> and CH<sub>4</sub> emissions from the whole Petit Saut system to the atmosphere during the 10 first years after impounding. Total carbon emissions (Figure 9c) were >0.35 million tons of carbon per year during the first 3 years and then decreased to ~100,000 t yr<sup>-1</sup> during the year 2001. CH<sub>4</sub>/CO<sub>2</sub> ratio (Figure 9d) decreased from 0.26 in 1994 to a relatively stable value around 0.1 since 1998. However, wet years like 2000 have higher CH<sub>4</sub>/CO<sub>2</sub> ratios, owing to higher degassing at the weir that skips the methane oxidation filter at the oxicle. Dry years like 2003 have a lower CH<sub>4</sub>/CO<sub>2</sub> ratio owing to lower CH<sub>4</sub> degassing flux but higher CO<sub>2</sub> diffusive flux. CO<sub>2</sub> emissions (Figure 9a) occurred in majority by diffusion from the reservoir surface (average 61%) and from the Sinnamary estuary (31%) with a small contribution of degassing at the outlet of the dam. CH<sub>4</sub> emissions from the estuary were generally less than 10% of the total (Figure 9b). CH<sub>4</sub> diffusion and bubbling from the reservoir surface were predominant only during the first year (respectively 40% and 44%). Since the year 1997, more than 60% of the CH<sub>4</sub> emissions occur as degassing at the weir. The 50% underestimation in the CH<sub>4</sub> diffusive flux when based on calculated data at station Roche Genipa (section 3.2.1.2) has a negligible impact on the total carbon emissions and a





**Figure 9.** Changes in total annual emissions from the whole Petit Saut system during 10 years since impounding: (a) CO<sub>2</sub> emissions, (b) CH<sub>4</sub> emissions, (c) total carbon emissions, and (d) CH<sub>4</sub>/CO<sub>2</sub> molar ratio. The diffusive flux is from the reservoir surface. The bubbling flux is from the reservoir shallow area. The degassing occurs at the outlet of the dam. The estuarine emissions were measured in 2003, were supposed as constant, and include the freshwater tidal river and the estuary; only half of the CO<sub>2</sub> estuarine emission was attributed to the presence of the Petit Saut reservoir.

moderate (less than 7%) impact on the total CH<sub>4</sub> emissions, which occur in majority through degassing. When reported to the flooded surface area, total gas fluxes and associated interannual variabilities ( $\pm$ SD) were  $192 \pm 13$  and  $92 \pm 36$  mmol m<sup>-2</sup> d<sup>-1</sup> for CO<sub>2</sub>, respectively for the periods 1994–1996 and 1997–2003 and  $42 \pm 10$  and  $11 \pm 17$  mmol m<sup>-2</sup> d<sup>-1</sup> for CH<sub>4</sub>, respectively for the same periods.

### 3.3. Net Carbon Budget of the Petit Saut System

[27] How long the flooded terrestrial OM remains in decomposition in reservoirs is a crucial question that still remains unsolved today [Delmas *et al.*, 2004; Tremblay *et al.*, 2004]. All the different OM pools (soils, litters, leaves, branches and trunks) are probably decomposed at very different speed and rates. In addition, this degradation might be very different in tropical, temperate and boreal reservoirs, owing to the different temperature and oxygen concentrations. The time course of gaseous emissions with reservoir age has always been established using data on different sites [Galy-Lacaux *et al.*, 1999; Saint Louis *et al.*, 2000; Rosa *et al.*, 2003]. As a consequence, little is known

on the long-term carbon budget of individual reservoirs and long-term fate of the flooded OM after impounding. In addition, in old reservoirs, OM carried by rivers and by surface runoff from surrounding land can become a significant source for CO<sub>2</sub> and CH<sub>4</sub> emissions. The 10 years CO<sub>2</sub> and CH<sub>4</sub> emissions calculated here offer the possibility to evaluate the carbon budget of the Petit Saut reservoir.

#### 3.3.1. Net Carbon Budget in 2003

[28] In order to compare quantitatively the carbon emissions with the carbon stock initially flooded in the Petit Saut reservoir, it is necessary to carefully consider all the different sources of carbon in the system, as well as the potential errors on each term. First, it must be checked whether riverine carbon loads to the reservoir significantly contribute to the atmospheric flux or not [Fearnside, 2002; Tremblay *et al.*, 2004]. In Table 5, we present a complete carbon budget of the Petit Saut system for the year 2003, based on our two last surveys. The budget includes for all carbon species the input by the rivers upstream and the output to the Atlantic Ocean downstream, as well as the gaseous emissions to the atmosphere. Among the calculated

**Table 5.** Carbon Budget of the Petit Saut Reservoir in 2003<sup>a</sup>

	Concentrations, $\mu\text{mol L}^{-1}$	Carbon Fluxes, $\text{t yr}^{-1}$
River input		
TOC	347	21,000
HCO <sub>3</sub> <sup>-</sup>	110	6,500
CO <sub>2</sub>	136	8,000
CH <sub>4</sub>	0.4	24
Total		35,500
Output to ocean		
TOC	460	28,000
HCO <sub>3</sub> <sup>-</sup>	131	8,000
CO <sub>2</sub>	263	16,000
CH <sub>4</sub>	0.1	6
Total		52,000
Emission to the atmosphere		
CO <sub>2</sub>		122,000
CH <sub>4</sub>		9,000
Total		131,000

<sup>a</sup>The river input is based on the May and December 2003 average concentrations at station Saut Takari Tanté upstream of the reservoir influence (Figure 1) and the 2003 average total watershed discharge of  $160 \text{ m}^3 \text{ s}^{-1}$ . The output to the ocean is based on May and December 2003 average concentrations at the most downstream freshwater station in the Sinnamary estuary (65 km downstream of the dam) and the same 2003 average discharge. The emissions to the atmosphere are from Table 4. Totals were rounded off at  $\pm 500$  for clarity.

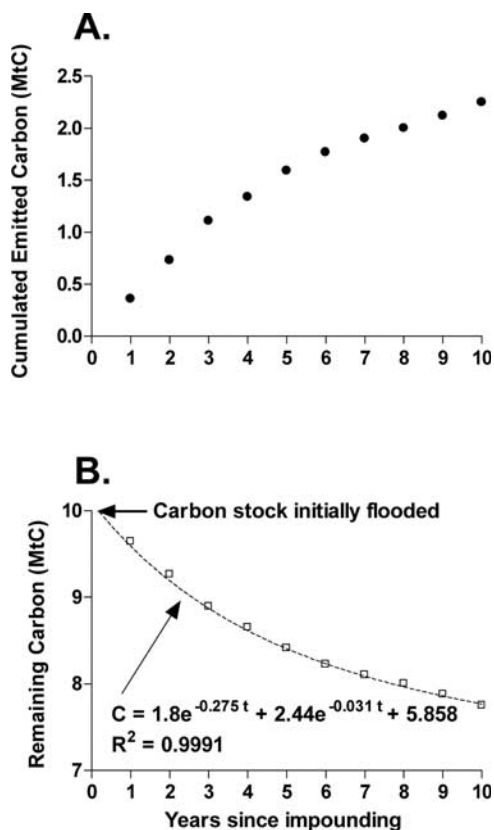
fluxes, the river input is the less robust estimate. First, the Sinnamary River contributes to only 50% of the total water flow and the assumption is made that other small tributaries have the same carbon concentrations, which is supported by the homogeneity of the reservoir watershed (primary forest). Second, more than 2 sampling period would be required to accurately quantify the input by rivers. Nevertheless, this estimate in Table 5 is, at least, the exact order of magnitude with a  $\pm 50\%$  confidence. Indeed, the majority of the carbon river influx is composed of TOC (Table 5), 90% of which is DOC. DOC concentrations show moderate temporal variations in these rivers: we measured DOC concentrations of  $302 \pm 35 \mu\text{mol L}^{-1}$  in the Sinnamary River at the upstream station Takari Tanté in 2003 ( $n = 2$ ) and Gadel *et al.* [1997] have measured DOC concentrations of  $350 \pm 57 \mu\text{mol L}^{-1}$  in the Coursibo and Sinnamary rivers in 1995 ( $n = 3$ ). Concerning the outflux to the ocean, the concentrations used for calculation (average of two sampling in 2003, 65 km downstream of the dam; Table 5), are very similar to the average concentrations measured 25 km upstream at station Pointe Combi (40 km downstream of the dam) during the 10 years' monitoring (Table 1). Although in Table 5, the Petit Saut system also appears as a net carbon exporter to the ocean, the input from the river may not be significantly lower than the output to the ocean. Nevertheless, Table 5 clearly indicates that, in 2003, the lateral carbon fluxes were  $\sim 3$  times lower than the carbon emissions to the atmosphere. This shows that 10 years after impounding, the flooded soil and biomass in the Petit Saut reservoir is still decomposing and contributing to most of the flux to the atmosphere. Degradation of OM from the watershed might also occur in the reservoir (by aerobic and anaerobic respiration and photo-oxidation), but this loss must be compensated by a carbon export to the ocean from the flooded OM pool. Primary production in the reservoir

is not considered in such “black box” budget because it simply changes the carbon from an inorganic to an organic form but does not affect the whole system budget. However, the production of highly labile OM in the reservoir by aquatic primary producers might fuel methanogenesis in the anoxic hypolimnion. That way, the CO<sub>2</sub> initially stored by aquatic primary production would be partly converted to CH<sub>4</sub>, making tropical reservoirs act as “methane factories” [Fearnside, 2004].

### 3.3.2. Long-Term Carbon Budget

[29] The cumulated amount of carbon emitted to the atmosphere by the whole system in 10 years (calculated using the data from Table 4) is  $\sim 2.2 \text{ Mt C}$ . This estimate is robust at about  $\pm 25\%$  since it takes into account all the emission terms of the system upstream and downstream of the dam and is based on a large data set that adequately integrates temporal variability. This carbon emission is very consistent with the predictions of Galy-Lacaux *et al.* [1999], who, on the basis of 3 years of measurements at Petit Saut, completed with data in four older Ivorian reservoirs, extrapolated a 3.2 Mt C emissions in 20 years. The 10 year data set thus confirms the conclusions by Delmas *et al.* [2001] concerning the global warming potential of the reservoir compared to other thermal alternatives.

[30] According to Galy-Lacaux *et al.* [1997], about 10 millions tons of carbon (Mt C) were flooded in Petit Saut. This estimation is based on an average carbon density of  $270 \text{ t ha}^{-1}$  [Carbon Dioxide Information Analysis Center, 1986; Olson *et al.*, 1983] and includes 62% of wood, 2% of leaves and 34% of soils and litters. In 10 years, the carbon Petit Saut reservoir has lost  $\sim 22\%$  of its initial flooded carbon. The atmospheric flux calculated here concerns however only the below-water part of the biomass. As pointed out by Fearnside [2002], an additional fraction of carbon may have reached the atmosphere through above-water losses. In Figure 10, we have plotted for the 10 years of observations the total carbon emitted to the atmosphere and remaining in place in Petit Saut (as discussed before, considering that the carbon loss is equivalent to the atmospheric emission). The obtained trend is typical for a kinetic decomposition of organic matter. On these data, we could fit a “double-G” model [Westrich and Berner, 1984] in which organic carbon is assumed to be the sum of a rapidly and a slowly decomposing fractions which both follow a first-order decay. This model fitted on the data with a  $R^2 > 0.999$  and gave a rapidly degradable fraction of 1.8 Mt C (18% of total) with a half-life of 2.5 years and a slowly degradable fraction of 2.4 Mt C (24%) with a half-life of 23 years. In this model, only 58% of the flooded carbon is nonbiodegradable and 64 years are needed to decompose 90% of the biodegradable carbon pool. However, although the parameters of the rapidly degradable fraction obtained with this model are intuitively consistent with the time course in Figure 9, the parameters for long-term decomposition are not statistically constrained. This means that the equation in Figure 10 cannot be used to predict the future carbon loss in the Petit Saut reservoir. The fact that 42% of the total soil and biomass is biodegradable in the model implies that a fraction of the wood (tree trunks and branches) would also decompose. If the same trend continues in the next decades,



**Figure 10.** (a) Cumulated carbon emitted as CO<sub>2</sub> and CH<sub>4</sub> to the atmosphere and (b) decrease in the carbon stock in 10 years. The dotted line is a “double-G” model that fit well to the data (see text).

it would confirm the results of *Campo and Sancholuz* [1998], who sampled submerged wood in a 46 years old tropical reservoir in Uruguay and showed it had lost 40% of its density.

#### 4. Conclusion

[31] This study is the first reconstruction of CO<sub>2</sub> and CH<sub>4</sub> emissions from a tropical reservoir during 10 years since impounding. This was possible owing to the large database available at Petit Saut. Seasonal variations were generally higher than interannual variation, which shows that estimations based on 1 to 2 surveys per year should be taken with caution. We also stress the importance of quantifying all emission terms. In particular, degassing at the aerating weir downstream of the dam was the predominant pathway to the atmosphere for CH<sub>4</sub>. This confirms the importance of quantifying this term, particularly in tropical stratified reservoirs with high CH<sub>4</sub> concentrations in the hypolimnion [Fearnside, 2004]. We also confirm the potential importance of tropical reservoirs as significant source of gases to the atmosphere, in particular for CH<sub>4</sub> [Saint Louis et al., 2000]. However, owing to the rapid decrease in CH<sub>4</sub> emissions, extrapolation at the global scale should be reassessed taking into account the trend reported here. In particular, the reservoir age is a key parameter that should

be taken into account. Finally, we show that, 10 years after impounding, the decomposition of the flooded soil and biomass is still the major contributor to the gas emission from the Petit Saut reservoir.

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