

Rainfall leads to increased pCO_2 in Brazilian coastal lakes

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Abstract. The variation of partial pressure of CO₂ (pCO₂), pH, salinity and dissolved organic carbon (DOC) in surface waters of 12 coastal Brazilian lakes was examined following periods of contrasting rainfall. Periods of high rainfall were followed by a large, almost 10 fold, increase in pCO₂ and a one unit decrease in pH in the lakes, whereas no consistent changes in DOC were observed. CO₂ emissions to the atmosphere from the Brazilian coastal lakes studied here were highly enhanced, on average, from $28.5 \pm 6.0 \text{ mmol C m}^{-2} \text{ d}^{-1}$ in dry periods to 245.3.1 ± 51.5 mmol C m⁻² d⁻¹ following periods of heavy rainfall. The increased inputs of pCO₂ following periods of high rainfall are believed to be derived from increased inputs of pCO₂ from groundwaters to the lakes.

1 Introduction

Although inland aquatic ecosystems occupy a small fraction of the continents (2 to 4%; Downing et al., 2006), these ecosystems can affect regional carbon balances, as they tend to support higher net carbon fluxes per unit area than those in surrounding terrestrial ecosystems (Cole et al., 2007; Tranvik et al., 2009). The disproportionate role of lakes in the carbon balance of landscapes derives from their role as recipients of a substantial fraction of the carbon produced within the watershed (Lennon, 2004; Sobek et al., 2005), and transported to lakes through surface runoff and groundwater flow, which are intensified following rainfall events (Schindler, 1978).

Terrestrial organic carbon, though relatively refractory (Hopkinson et al., 1998), plays an important role in lakes,



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where it partially supports aquatic food webs (Pace et al., 2004), metabolism (Cole et al., 2000) and contributes to the prevalent carbon dioxide (CO₂) supersaturation of lake waters (Sobek et al., 2005). Allochthonous inputs of CO₂ can also contribute to maintain the partial pressure of carbon dioxide (pCO_2) above equilibrium with the atmosphere in aquatic ecosystems (Raymond and Cole, 2003). CO₂ enrichment of lake waters may closely follow the precipitation pattern (Rantakari and Kortelainen, 2005), possibly reflecting the associated inputs of surface and groundwater waters containing high concentrations of terrestrial organic and inorganic carbon (cf. Cole et al. 2007).

Tropical coastal lakes in Brazil are typically small, shallow, and broadly distributed in watersheds with important components of Restinga, the characteristic vegetation of Atlantic Tropical Forest, occupying extensive areas of sand plain along the coastline. This vegetation is a major source of carbon to these lakes, which waters are often highly colored (Amado et al., 2007). Recent analyses have shown Brazilian lakes to be highly supersaturated in CO₂ (Marotta et al., 2009), supported by high inputs of terrestrial-derived carbon to these lakes. Groundwater plays an important role in the water budget and inputs of materials to Brazilian coastal lakes, a role that can be intensified by high rainfall, due to low water retention by the sandy soils in their watershed (Farjalla et al., 2002). Indeed, pCO_2 in Brazilian coastal lakes have been shown to vary greatly and synchronously over time (Marotta et al., 2010), suggesting weather-control of this property. Because temperature shows little temporal variability in this tropical region, rainfall, which may affect the discharge of groundwater and the associated inputs of carbon to the lakes and that vary greatly over time, may be the driver behind this synchronous pCO_2 variability. However, the role of rainfall in accounting for variability in CO₂ in these ecosystems has not yet been tested.

We examined here the variability in surface pCO_2 , pH, salinity and DOC in a series of coastal Brazilian lakes to test the hypothesis of a relationship between variability in pCO_2 and that in rainfall in these lakes.

2 Material and methods

2.1 Study area

The coastal lakes studied are located in Rio de Janeiro state, characterized by warm temperatures, ranging from a minimum monthly average temperature of 20.7 °C in July to a maximum of 26.2 °C in February (INMET, 1992). The mean minimum and maximum rainfall are typically observed in August (38 mm) and December (182 mm; INMET, 1992), but show high inter-annual variation (Carmouze et al., 1991).

The 12 studied lakes (Fig. 1) are situated in the National Park of Restinga de Jurubatiba, one of the most important protected areas with coastal lakes in Brazil. This conservation area has many small lakes separated from the sea by sandbars along the shoreline, which may open up during episodic events, including extreme rainfall and sea storms, or human intervention.

The studied lakes stretch along a 40 Km strip of coastline (22°00' and 22°23' S and 41°15' and 41°45' W; Fig. 1), are small (area $<5 \text{ km}^2$; Table 1), shallow (maximum depth varying from 0.8 to 4.0 m), and affected by saltwater intrusions resulting in brackish-saline waters (salinity range from 2 to 33). Most lakes of this region do not have surface freshwater inlets but are fed by groundwater. Rainfall exerts an important dynamic control on lake depth through groundwater inputs, which contribute high DOC (Suzuki et al., 1998). and CO₂ inputs, as groundwaters are highly supersaturated in CO₂ (Suzuki et al., 1998).

2.2 Study Design

The general sampling strategy involved a series of sampling events between 2003 and 2006, each involving measurements along 24 h cycles to characterise daily variability in pCO_2 and preceded by contrasting weekly-accumulated rainfall. Sampling effort at Lake Carapebus was more intense, with pCO₂, pH, salinity and temperature analyzed 9 times over two consecutive daily cycles (06:00 p.m., 06:00 a.m., 10:00 a.m., 02:00 p.m., 06:00 p.m. the following day) and only once for DOC concentrations in each sampling event (N = 6 samplings events between 2003 and 2004). Two sampling stations, characterized by a similar oligomesotrophic status during this period (about $0.8 \,\mu mol \, L^{-1}$ of total phosphorus and $5 \,\mu g \, L^{-1}$ of chlorophyll-*a* concentrations), were analyzed in this lake. A station colonized by submerged aquatic plants (macrophyte covered), mainly Potamogetum stenostachys with a mean \pm SE standing crop of 1430 ± 200 g m⁻² (N = 21 quadrats between 2004 January and July), and a station devoid of macrophytes (open waters).



Fig. 1. Location of the studied lakes (1) Carapebus, (2) Garças, (3) Peri-peri 1, (4) Peri-peri 2, (5) Maria Menina, (6) Robalo, (7) Visgueiro, (8) Catingosa, (9) Pires, (10) Preta, (11) Barrinha and (12) Casa Velha.

Table 1. General characteristics of the studied lakes.

Lakes	Total P*	Colour*	Area
	$(\mu mol L^{-1})$	(at 430 nm)	(km ²)
Garças	0.4–0.5	0.026-0.089	0.42
Peri-peri 1	0.7–0.9	0.036-0.040	0.14
Peri-peri 2	0.8 - 1.0	0.080-0.138	0.83
Maria Menina	0.5 - 0.7	0.019-0.089	0.24
Robalo	0.4–0.6	0.014-0.026	0.85
Preta	0.2 - 0.7	0.217-0.307	2.19
Pires	0.4 - 1.0	0.071 - 0.075	0.92
Catingosa	0.5 - 0.7	0.013-0.049	0.09
Visgueiro	0.3-1.0	0.021-0.037	1.18
Casa Velha	0.2 - 0.5	0.044-0.068	0.54
Barrinha	0.8 - 1.1	0.049–0.087	0.24
Carapebus	0.6–1.1	0.009-0.028	4.33

* Range considering the sampling events in each lake.

A total of 11 additional oligo to mesotrophic (total phosphorus below 1.1 µmol L⁻¹, Table 1) coastal lakes were sampled in the National Park of Restinga de Jurubatiba. pCO_2 , pH, salinity and temperature in surface waters were simultaneously analyzed 4 times over a daily cycle (06:00 p.m., 06:00 a.m., 12:00 a.m. and 06:00 p.m. the following day) and only once per daily cycle for DOC concentrations, in each of two sampling events preceded by dry and rainy periods, both in 2006.

2.3 Analytical Methods and pCO₂ Calculations

Surface water samples were immediately analyzed for pH and alkalinity. Temperature and salinity were measured in situ with a calibrated YSI-30 Thermosalinometer. At the laboratory, pre-filtered (0.7 µm, Whatman GF/F) water samples were acidified to pH <2.0 and analyzed for DOC concentrations using high-temperature catalytic oxidation on a Shimadzu TOC-5000 Analyzer. CO2 concentrations and dissolved inorganic carbon concentrations (DIC) were calculated from pH and alkalinity measurements after correction for temperature, altitude and ionic strength following Cole et al. (1994) and Weiss (1974). pH was measured with a precision of 0.01 pH units using a calibrated Analion PM 608 pH meter and total alkalinity by Gran's titration (APHA, 1992). pCO_2 was calculated using Henry's law, as the ratio between the CO₂ concentration and Henry's constant for this gas at a given temperature and salinity.

Data on rainfall were obtained from the Brazilian Aerospace Technical Center (CTA, São José dos Campos). Rainfall data were aggregated as the cumulative rainfall in the week preceding each sampling event, encompassing the time lag between rainfall events and hydrological inputs as well as the time scales for responses of lake metabolism to perturbations (Staehr and Sand-Jensen, 2007).

Log-transformed data showed significant Gaussian distribution (Kolmogorov-Smirnov, p < 0.05), homogeneity of variances (Bartlett, p > 0.05) and significant pairing (F test, p < 0.05). Hence, sampling events were compared using paired parametric tests with a significance of p < 0.05 (Zar, 1996). We used paired t-test to compare two data sets or repeated measures one-way ANOVA followed by Tukey-Kramer test for multiple comparisons. All statistics were calculated using the software Graphpad Prism 4.0. Daily means of pCO_2 were calculated in each sampling event and lake (two daily cycles in Lake Carapebus and only one in other lakes).

3 Results

Daily variability in pCO_2 (coefficient of variation = 68%) and pH (6.5 to 7.5) was substantial, and comparable in magnitude to as variability across lakes after dry or rainy periods (coefficient of variation = 75% and 6.5 to 8.0 respectively). However, the daily range between minimum and maximum pCO_2 and pH were much larger, increasing from 3.5 to 50 fold and from 0.5 to 8 fold, respectively, following rainy periods in all studied lakes, except in Lake Carapebus, where the daily range in pH was comparable between both rainy and drier periods. The mean increase in maximum daily values of pCO_2 following high weekly-accumulated precipitation was about 6 fold (paired t-test, p < 0.0001), well above that experienced by minimum daily pCO_2 values (about 85%; p < 0.05, paired t-test).



Fig. 2. Accumulated precipitation in the week preceding the sampling (full squares) and average DOC concentrations (**a**), pCO_2 (**b**), salinity (**c**) and pH (**d**) in open waters (triangles) and macrophyte-covered (crosses) waters of Lake Carapebus along the studied period. Bars indicate standard errors and at least a letter shared by different dates indicates no significant differences (Tukey-Kramer, p < 0.05) for pH and pCO_2 among stations and sampling events.

The increase in weekly-accumulated rainfall at Lake Carapebus (Fig. 2) was coupled to great pCO_2 enrichments (about 11 fold in the macrophyte covered station and 4 fold in the open water station), two-fold higher DOC concentrations, a decrease of 50% in salinity and a reduction in pH by 1.0 unit (Tukey-Kramer and paired t-test, p < 0.05; Fig. 2 and Table 2). Indeed, there were strong positive relationships of lake pCO₂ and DOC with respect to weekly-accumulated rainfall, which was negatively correlated with pH and salinity, in both Lake Carapebus stations (linear regression, p <0.05; Fig. 3). High rainfall also homogenised pCO_2 within Lake Carapebus, as the high intra-ecosystem heterogeneity for pCO_2 (pCO_2 about 4 times higher in the open water compared to the station with submerged macrophytes) was reduced to a non-significant difference following intense rain. In contrast, the extent of variability in water temperature

Lakes	7-days precipitation (mm)	pCO ₂ (µatm)	DOC $(mg L^{-1})$	pH (US)	Salinity
Garças	1.2	871	17.7	8.4	33.2
	38.7	4419	23.0	7.8	32.2
	<i>Ratio</i>	5.07	1.30	- 0.6	0.97
Peri-peri 1	1.2	552	18.8	8.5	25.7
	38.7	8389	34.8	7.4	12.9
	<i>Ratio</i>	15.20	<i>1.30</i>	- 1.1	0.50
Peri-peri 2	1.2	1927	38.0	7.8	10.6
	38.7	6644	43.8	7.7	14.0
	<i>Ratio</i>	<i>3.45</i>	1.15	- 0.1	<i>1.32</i>
Maria Menina	1.2	1889	35.9	8.0	18.1
	38.7	6387	42.7	7.7	15.8
	<i>Ratio</i>	<i>3.3</i> 8	1.19	0.3	0.87
Robalo	1.2	1611	43.0	8.3	25.8
	38.7	13736	38.3	7.6	20.0
	<i>Ratio</i>	8.53	0.89	- 0.7	0.77
Preta	1.2	2115	39.0	7.6	3.3
	38.7	5950	42.0	7.3	4.5
	<i>Ratio</i>	2.81	1.08	- 0.3	1.36
Pires	1.2	1564	35.5	8.0	7.0
	38.7	5505	34.5	7.5	3.5
	<i>Ratio</i>	3.52	0.97	- 0.5	0.50
Catingosa	1.2 38.7 <i>Ratio</i>	3446 20037 5.81	Nc 40.1	7.8 7.1 - 0.7	20.6 20.5 0.99
Visgueiro	1.2	2643	37.6	8.0	21.8
	38.7	16672	37.8	7.2	22.1
	<i>Ratio</i>	6.31	1.01	- 0.8	1.01
Casa Velha	1.2	361	41.2	8.7	5.0
	38.7	1153	40.9	8.3	7.0
	<i>Ratio</i>	<i>3.19</i>	0.99	- 0.4	1.40
Barrinha	1.2	1167	42.9	8.2	4.9
	38.7	2684	43.1	7.8	6.5
	<i>Ratio</i>	2.30	1.00	- 0.4	1.32
CAR_ Macrophytes	0.0–6.7	386	9.1	7.9	6.0
	34.6–67.6	4203	17.9	6.8	2.2
	<i>Ratio</i>	10.90	1.98	- 1.1	0.37
CAR_ Open	0.0–6.7	1609	8.1	7.4	6.2
	34.6–67.6	5905	16.6	6.6	2.4
	<i>Ratio</i>	<i>3.67</i>	2.05	- 0.8	0.39

Table 2. Daily-average values of pCO_2 , DOC, pH and salinity after periods of high and low weekly-accumulated precipitation events for each studied lake, and the ratio of high rainfall to low rainfall values.

nc = not collected.



Fig. 3. The relationships between mean pCO_2 , DOC, salinity and pH with accumulated precipitation in the week preceding the sampling in the macrophyte covered (Figs. 3a, b, c, d) and open water (Figs. 3e, f, g and h) stations at Lake Carapebus. Solid lines represent significant linear regressions (p < 0.05).

was independent of the previous rainfall (linear regression, p > 0.05).

A pattern towards sharply increasing pCO_2 and decreasing pH with increasing rainfall was also evident considering all remaining lakes sampled, with pCO_2 increasing by 3 to 15 fold and pH decreasing from 0.1 to 1.1 units after rainy periods (Table 2). Lake pCO_2 and pH were strongly correlated, within sampling events and for both sampling events combined (linear regression, p < 0.05; Fig. 4). In contrast, changes in DOC concentrations did not follow a consistent trend with rainfall across the 11 lakes sampled simultaneously (Table 2). There was no significant relationship between pCO_2 and DOC within each sampling event or for both sampling events combined (linear regression, p > 0.05; Fig. 4).

Salinity fluctuations were not synchronous among the lakes in both sampling events, and changes in salinity were independent of those in pCO_2 among lakes (linear regression, p > 0.05; Fig. 4). However, lakes with a higher decline in salinity following intense rainfall (like Peri-peri1 and Robalo) showed higher pCO_2 enrichment in the rainy compared to the drier period (Table 2). Indeed, the observed changes in pCO_2 in the 11 lakes sampled between the drier and rainier sampling events were significantly correlated with changes in salinity and pH (positive and negative, respectively), but not with fluctuations in DOC (Table 2 and Fig. 5, linear regression, significant p < 0.05). Another important driver of lake pCO_2 , water temperature, remained relatively uniform between both sampling events $(21.5 \pm 0.3 \,^{\circ}\text{C}, \text{ mean } \pm \text{SE})$ and was not also significantly related to the large differences of CO₂ saturation observed in the lakes studied (t-test, p > 0.05).



Fig. 4. The relationships of pCO_2 with (a) DOC, (b) salinity and (c) pH in 11 lakes sampled simultaneously within drier (crosses) and rainier (full triangles) sampling events. Only pH showed significant linear regressions (p < 0.05) with pCO_2 in each period ($pCO_2 = -2214 \text{ pH} + 19,620$, $R^2 = 0.61$ for the drier one; $pCO_2 = -13,640 \text{ pH} + 111,800$, $R^2 = 0.66$ for the rainier one), and combining both ($pCO_2 = -9823 \text{ pH} + 82,090$, $R^2 = 0.60$).

4 Discussion

Most of the lakes examined had waters supersaturated in CO_2 , acting therefore as CO_2 sources to the atmosphere in all sampling events. Only two lakes showed daily mean pCO_2 below atmospheric equilibrium (about 380 µatm). The lowest pCO_2 values were observed in the station colonized by submerged macrophytes in Lake Carapebus, consistent with the role of submerged vegetation as CO_2 sinks in natural waters (Krause-Jensen and Sand-Jensen, 1998).

Increased, weekly-accumulated, precipitation prompted considerable increase in pCO_2 and the amplitude of daily pCO_2 variability, despite minor changes in water temperature. Higher amplitude of diel CO₂ changes in lakes (Staehr and Sand-Jensen, 2007) might reflect increases in the metabolism of lake ecosystems (Richey et al., 2002). Here, the strong increase in mean and maximum daily pCO_2 values, well above those in the minimum daily pCO_2 values, resulted in broad daily pCO_2 oscillations following periods of high rainfall. This result confirms that intense rainfall may be correlated with CO₂ enhancements in small tropical lakes even one order of magnitude higher than previously reported for large boreal lakes (Rantakari and Kortelainen, 2005).

The positive relationship of pCO_2 with DOC, and negative relationship with pH and salinity, following periods of high rainfall at Lake Carapebus is consistent with evidence that terrestrial freshwater inputs enhance CO₂ production in lake ecosystems (Cole et al., 1994; Cole et al., 2007). It also confirms the potential role of rainfall on the inputs of terrestrial organic substrates to aquatic CO₂ production by respiration in lakes (Rantakari and Kortelainen, 2005).

Groundwater inputs have been shown to be important drivers of changes in coastal tropical lakes elsewhere (HerreraSilveira, 1996), and are a major vector of inputs of humic acids to the lakes studied here (Farjalla et al., 2002). The Restinga vegetation in the lake watershed provides abundant organic carbon to the soils, which can be entrained to groundwater along with the CO_2 derived from respiration of this organic carbon during periods of intense rainfall and eventu-



Fig. 5. The relationships of the variation rates, comparing the rainier sampling event to drier one, of pCO_2 with (Fig. 4a) DOC, (Fig. 4b) salinity and (Fig. 4c) pH in the 11 lakes sampled simultaneously. Solid lines represent the significant fitted regression equations (p < 0.05) for changes in salinity (variation rate in $pCO_2 = -7.4$ variation rate in salinity + 12.8, $R^2 = 0.41$) and pH (variation rate in $pCO_2 = -11.5$ variation rate in pH -0.8, $R^2 = 0.75$).

ally delivered to the lake (Marotta et al., 2010). Analysis of stable carbon isotopic signatures has also revealed that DOC in the lake waters in the study region largely represent allochthonous carbon from terrestrial sources delivered to the lake by groundwaters (Marotta et al., 2010). pCO_2 values in groundwaters in the study region can reach very high values, e.g., up to 70,000 µatm or about 180 times higher than the equilibrium with the atmosphere (Suzuki et al., 1998), so that groundwater inputs can support large pCO_2 emissions when ventilating in lake waters. Indeed, the high CO₂ in groundwaters, along with the high concentration of terrestrial humic acids, account for the low pH of groundwaters (Jones and Mulholland, 1998), explain the general reduction in pH observed in the lakes studied following periods of intense rainfall.

The coupling between high rainfall and high pCO_2 and a decrease in pH and salinity observed in Lake Carapebus was consistent with the changes observed in the 11 lakes sampled simultaneously after rainy events. In contrast, the lack of a significant relationship between the variability in pCO_2 and DOC in these lakes indicates that strong pCO_2 enrichment with increasing rainfall is not always accompanied by an increase in DOC. Indeed, there was no significant relationship between lake DOC and pCO_2 in the lakes sampled here, in contrast to the general relationship observed at the global scale (Sobek et al., 2005). Yet, the broader daily variation in pCO_2 following periods of intense rainfall indicates that ecosystem metabolism is enhanced following rainfall.

Our results confirmed the hypothesis and previous findings on the high pCO_2 variability for tropical inland waters, suggesting a possible controlling role of rainfall. Here, large pCO_2 enhancements in lake waters following increases in weekly-accumulated rainfall were even comparable to the variation observed among lakes (Marotta et al., 2009) and within lakes (Marotta et al., 2010), or that related to seasonal (Richey et al., 2002) and interannual (Marotta et al., 2010) changes in inland waters at low latitudes. The results presented demonstrate an important role of rainfall, and the subsequent groundwater inputs, in generating variability in pCO_2 in the Brazilian coastal lakes studied, with a large increase in pCO_2 , and a decline in pH, following higher rainfall. The diel variability reported also suggests that intense rainfall also stimulates ecosystem metabolism conducive to broad variability in pCO_2 along the day. Whereas groundwater contributes relatively small amounts of CO₂ directly to the atmosphere (Cole et al., 2007), the ventilation of the high CO₂ in groundwater may support intense CO₂ emissions when entering lakes. Estimates of the CO₂ emissions to the atmosphere from these lakes were derived following calculations in Cole and Caraco (1998), a pCO_2 in equilibrium with the atmosphere of 380 µatm and the mean global wind velocity reported over land of $3.28 \,\mathrm{m \, s^{-1}}$ (Archer and Jacobson, 2005). CO₂ emissions from Brazilian coastal lakes studied here were enhanced, on average, almost 10 fold following intense rainfall, from an average $(\pm SE)$ of $28.5 \pm 6.0 \text{ mmol C m}^{-2} \text{d}^{-1}$ in drier periods to $245.3 \pm 51.5 \text{ mmol C m}^{-2} \text{d}^{-1}$ following heavy rain. These results suggest that precipitation and subsequent ventilation of groundwater CO₂ at the lake-atmosphere interface may provide a conduit to deliver CO₂ ultimately resulting from soil respiration to the atmosphere.

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