Thermal enhancement of gas transfer velocity of CO$_2$ in an Amazon floodplain lake revealed by eddy covariance measurements

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In November 2011, the partial pressures of carbon dioxide (pCO$_2$) in water and air in a floodplain lake of the Amazon River in Brazil were 800±75 and 387±8 ppmv, respectively. Turbulent CO$_2$ fluxes from the lake measured with eddy covariance ranged from 0.05 to 2.2 μmol m$^{-2}$ s$^{-1}$. The corresponding gas transfer velocities $k_{500}$ ranged from 1.3 to 31.6 cm h$^{-1}$, averaging 12.2±6.7 cm h$^{-1}$. At moderate to high wind speed, $k_{500}$ increased with wind speed, with values above parameterizations for other lake ecosystems. During the prevailing tropical low wind speed (below 2.7 m s$^{-1}$) and high insolation conditions, unexpected high $k_{500}$ values (up to 20 cm h$^{-1}$) were obtained and correlated with latent heat and sensible heat fluxes. In Amazonian open lakes, owing to long quiescent periods of low wind speed but extremely high daytime insolation and heat fluxes, thermal enhancement makes time-integrated gas transfer velocities four to five times higher than those computed from classic wind parameterization.


1. **Introduction**

Inland waters have been recognized as a globally significant source of carbon dioxide (CO$_2$) to the atmosphere that deserves detailed scientific investigation and better quantification. The CO$_2$ flux from waters to the atmosphere is proportional to the water-air CO$_2$ concentration gradient and the gas transfer coefficient $k$ [Cole and Caraco, 1998]. The magnitude of $k$ is controlled by the near-surface turbulence at the air-water interface [Zappa et al., 2003]. In lakes and oceans, $k$ is parameterized as a function of the wind speed, the dominant driver of turbulence [Cole and Caraco, 1998; Ward et al., 2004]. In macrotidal estuaries, the turbulence generated by tidal currents is particularly significant, leading to higher $k$ estimations in such environments [Zappa et al., 2003; Abril et al., 2009]. The fetch according to the wind exposure also modulates the speed of $k$ variations, making $k$-wind speed relationships site specific [Abril et al., 2009]. In lakes, other factors than wind that can modulate the turbulence at the aqueous mass boundary layer are thermal factors, such as evaporation and buoyancy fluxes [Eugster et al., 2003; MacIntyre et al., 2010]. Water convective mixing associated with heat losses at the air-water interface creates turbulence that leads to an enhancement of $k$. On the contrary, water stratification associated with heat gains tends to suppress turbulence and can have the opposite effect on $k$ [Eugster et al., 2003; Jonsson et al., 2008; MacIntyre et al., 2010].

[3] Concomitant and accurate measurements of the partial pressure of CO$_2$ (pCO$_2$) and the CO$_2$ flux ($F_{CO_2}$) at the air-water interface are required to directly estimate the gas transfer coefficient ($k$) in aquatic systems. Water pCO$_2$ can be measured at sufficient temporal and spatial resolutions using equilibrators. CO$_2$ fluxes at the air-water interface can be measured using floating chambers [Guérin et al., 2007; Abril et al., 2009], although this technique may perturb the boundary layer and increase the turbulence at the water surface, especially over sheltered lakes during low wind speed conditions. The eddy covariance (EC) technique is an appropriate noninvasive method for CO$_2$ flux measurements in aquatic systems based on measurements of the covariance between fluctuations in the vertical wind velocity and CO$_2$ mixing ratio at very high frequencies [Aubinet et al., 2000]. The area sampled by this technique, referred to as the footprint, ranges from 100 m to several kilometers depending on measurement height, surface roughness, and atmospheric stability. The EC requires important qualitative and quantitative analyses as well as corrections based on the physical and theoretical backgrounds underlying the method. Together with water pCO$_2$ measurements, the EC allows computation of $k$ on time scales that are short enough to resolve short-term variability occurring in highly dynamic systems. Few studies have applied the EC in boreal, temperate, and tropical lakes; these studies reported CO$_2$ fluxes alone [Anderson et al., 1999; Eugster et al., 2003], CO$_2$ fluxes with water as well as latent (LE) heat and sensible (H) heat fluxes [Morison et al., 2000; Vesala et al., 2006], or CO$_2$ fluxes with simultaneous water and air pCO$_2$ to derive $k$ values [Guérin et al., 2007; Jonsson et al., 2008]. The EC has also been used to compare the productivity of macrophyte meadows in an Amazonian floodplain during the aquatic and terrestrial phases [Morison et al., 2000].
In the Amazon basin, total CO₂ emission from the flooded area to the atmosphere has been estimated at 0.47 Pg C yr⁻¹, which is 10 times higher than the riverine organic carbon flux to the Atlantic Ocean (Richey et al., 2002). A large part of this flux comes from floodplains, where waters are supersaturated in CO₂. To derive this flux, the applied gas transfer velocity was 2.7 ± 1.0 cm h⁻¹, which corresponds to a wind speed parameterization in temperate lakes. In this study, we investigated the factors that control k in floodplain lakes of the Amazon River. The variations of k values derived from EC measurements are discussed as a function not only of wind speed but also of latent heat and sensible heat fluxes. We conclude that because of thermal effects under tropical climatic conditions, k parameterizations as a function of wind speed alone would underestimate gas transfer intensity.

2. Materials and Methods

2.1. Study Site

Data were gathered from a floodplain lake of the Amazon River in northern Brazil (Figure 1a) called Canaçari (3°5′S and 2°49′S–58°22′W and 58°9′W), which is located on the left edge of the Amazon River, 200 km downstream of the city of Manaus. Lake Canaçari is a relatively homogeneous open floodplain lake with a surface area of 450 km² that does not dry by more than 20% in surface area at the lowest water levels of the Amazon River. The lake receives in great majority “white” turbid waters from the Amazon River, whereas clear waters from the Urubu River that drain the northern local basin are generally bypassed to the northeast. The field cruise occurred during the dry season in November 2011 at relatively low water levels.

2.2. Eddy Covariance and Equilibrator Techniques and Calculations

Turbulent fluxes of CO₂, latent heat, and sensible heat and associated parameters were measured using an EC system positioned at the edge of the lake (2°57′53.76′′S, 58°17′6.12′′W; Figure 1a) over 4 days from 19 to 22 November 2011. The station was selected according to the dominant wind direction so that it receives air masses flowing over the lake. Our EC system (Polsenaere et al., 2012) was set on a mast at a height of 4.6 m above the water and consisted of a sonic anemometer (model CSAT3, Campbell Scientific Inc., Logan, Utah, USA) that measured the three wind speed components (m s⁻¹), the wind direction, and the sonic air temperature (°C) as well as an infrared gas analyzer (model LI-7500, Licor Inc., Lincoln, Nebraska, USA) that measured CO₂ and H₂O concentrations (mmol m⁻³) and atmospheric pressure (kPa). Analogue output signals from these fast-response instruments were sampled and digitized at the rate of 20 Hz. Additionally, a quantum sensor (SKP215, Skye Instruments, Llandrindod Wells, UK) and a meteorological transmitter (model WXT510, Vaisala Inc., Finland) were used to measure, respectively, photosynthetically active radiation (PAR; μmol m⁻² s⁻¹) and weather parameters every minute. Data were recorded by a central acquisition system (model CR3000, Campbell Scientific Inc.).

Concomitantly, the pCO₂ in lake waters around the EC station was measured every minute from a small boat by a custom-made equilibrator system adapted from Abril et al. [2006]. An infrared gas analyzer (LI820, Licor Inc.) measured the pCO₂ in dry air that was equilibrated with lake water. To assess the spatial heterogeneity of water pCO₂ in the lake within the footprint of the EC station, different recording tracks were realized according to wind directions (Figure 1). Continuous recording of water temperature was performed with a YSI-6920 probe at 0.7 m below the surface.

Data processing and quality control protocols for the EC technique were performed as described by Polsenaere et al. [2012]. Briefly, fluxes were computed with an averaging time of 10 min and data were processed using the EdiRe software from the University of Edinburg applying the following steps: (1) spike removal, (2) coordinate rotations, (3) linear detrending, (4) time lag corrections, (5) high-frequency corrections, and (6) (co)spectral analysis. According to data quality control protocols, two main statistical tests were applied, the steady state test and a test based on the integral turbulence characteristics of wind components and temperature, according to Foken et al. [1991]. At the end, 62%, 60%, and 64% of CO₂, LE heat, and H heat flux data, respectively, were retained in the acquired data set, corresponding to “high-quality data” (Foken, 2003).

The gas transfer velocity k was calculated using the air-water flux formulation \( F_e = \frac{\alpha k}{\Delta p CO_2} \), where \( F_e \) is the mean vertical CO₂ exchange at the air-water interface measured by EC, \( \alpha \) is the CO₂ solubility coefficient, and \( \Delta p CO_2 \) is the gradient between mean water and air pCO₂ measured by the equilibrator and the EC, respectively. The k value was then normalized to a Schmidt number of 600 (Sₘ = 600, for CO₂ at 20°C) as described by Guérin et al. [2007]. Mean \( k_{600} \) values were calculated over each 10 min period. We evaluated uncertainties in \( k_{600} \) associated with the heterogeneity of water pCO₂ within the EC footprint by considering the minimum and maximum \( k_{600} \) values corresponding to the minimum and maximum water pCO₂ values over each 10 min measurement. Finally, wind speed values measured at the mast height were normalized to a 10 m height according to Amoroc and DeVries [1980].

The available fetch represented by lake water always ranged from at least 1500 m (the closest land edge to the EC mast, in the west). Unstable atmospheric conditions prevailed over the lake as endorsed by always negative Monin-Obukhov parameter values measured over the 4 days (Z/L: −0.48 ± 0.62, −3.16 to −0.07). It is generally accepted that the relative height/footprint ratio must be 1:100 and 1:300 for unstable and stable atmospheric conditions, respectively (Leclerc and Thurtell, 1990). According to the height of EC sensors, we estimated the EC footprint to be 500 m. Thus, water pCO₂ measurements were always performed well within the footprint area of the EC measurements and according to the direction of prevailing winds (Figure 1).

3. Results

Air and water temperatures were generally close to each other with means of 30.1°C ± 1.4°C and 31.0°C ± 1.2°C, respectively (Table 1). However, water temperatures were always slightly higher than air temperatures during daytime measurements. No rain occurred during the study, and PAR values ranged from 20 to more than 2000 μmol m⁻² s⁻¹. The lowest PAR values occurred during morning measurements and the highest values in the middle of the afternoon.
Figure 1. (a) Location of Lake Canaçari showing the position of the EC measurement system (asterisk). (b–h) Boat tracks in the lake in front of the EC station showing the measured water pCO$_2$ (ppmv) with the equilibrator. Indicated times in minutes are from the beginning of the initial recording, which lasted about 2 h 20 min each. Wind rose plots during measurement periods are indicated in gray. Each measurement series lasted from 2 h to 2 h 30 min. Data were obtained on 19 November 2011 (from 09:30 until 11:30; Figure 1b), 20 November 2011 (from 09:50 until 12:00 in Figure 1c and from 15:30 until 17:40 in Figure 1d), 21 November 2011 (from 10:10 until 13:10 in Figure 1e and from 14:40 until 17:10 in Figure 1f), and 22 November 2011 (from 10:00 until 12:10 in Figure 1g and from 14:10 until 15:40 in Figure 1h).
Table 1. Summary of the Data Set with Averages, Standard Deviations (SDs) and Ranges (Minimum and Maximum), and Numbers of Measurements (N)

<table>
<thead>
<tr>
<th></th>
<th>Mean ± SD</th>
<th>Min. to Max.</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>T_a (°C)</td>
<td>30.1 ± 1.4</td>
<td>26.6–32</td>
<td>99</td>
</tr>
<tr>
<td>T_s (°C)</td>
<td>31.0 ± 1.2</td>
<td>28.7–34.6</td>
<td>92</td>
</tr>
<tr>
<td>PAR (μmol m⁻² s⁻¹)</td>
<td>1348 ± 758</td>
<td>22–2272</td>
<td>99</td>
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<tr>
<td>Wind direction (°)</td>
<td>94 ± 34</td>
<td>8–198</td>
<td>100</td>
</tr>
<tr>
<td>Uₐ₀ (m s⁻¹)</td>
<td>3.1 ± 1.7</td>
<td>0.6–6.5</td>
<td>100</td>
</tr>
<tr>
<td>Air pCO₂ (ppmv)</td>
<td>387 ± 8</td>
<td>372–403</td>
<td>99</td>
</tr>
<tr>
<td>Water pCO₂ (ppmv)</td>
<td>800 ± 75</td>
<td>666–1030</td>
<td>85</td>
</tr>
<tr>
<td>ΔpCO₂ (ppmv)</td>
<td>415 ± 72</td>
<td>277–627</td>
<td>85</td>
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<tr>
<td>Fₜ (μmol m⁻² s⁻¹)</td>
<td>0.59 ± 0.39</td>
<td>0.05–2.2</td>
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<tr>
<td>LE (W m⁻²)</td>
<td>198.3 ± 74.1</td>
<td>62.1–378.9</td>
<td>58</td>
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<tr>
<td>H (W m⁻²)</td>
<td>19.1 ± 10.4</td>
<td>4.9–49.1</td>
<td>65</td>
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<tr>
<td>k₆₀₀ (cm h⁻¹)</td>
<td>12.2 ± 6.7</td>
<td>1.3–31.6</td>
<td>44</td>
</tr>
</tbody>
</table>

*Shown are air and water temperatures (T_a and T_s); photosynthetically active radiation (PAR); wind speed at a 10 m height above the water surface (Uₐ₀); air and water pCO₂ and water-air gradient; turbulent CO₂, latent heat, and sensible heat fluxes (Fₜ, LE, and H); as well as gas exchange coefficient normalized to the Schmidt number of CO₂ at 20°C in freshwater (k₆₀₀).*

[Figure 2.](#) Complete data set acquired during the seven measurement periods. (a) Wind speed at a 10 m height above the water surface (mean Uₐ₀, black inverted triangles, m s⁻¹) and photosynthetically active radiation (PAR; squares, μmol m⁻² s⁻¹). (b) Water pCO₂ (black circles and standard deviations, vertical lines) and air pCO₂ (open circles). (c) Water-air CO₂ fluxes (mean Fₜ, black circles, μmol m⁻² s⁻¹). (d) Latent heat fluxes (mean LE, black squares, W m⁻²). (e) Sensible heat fluxes (mean H, black triangles, W m⁻²). (f) Gas transfer velocity normalized to the Schmidt number of CO₂ at 20°C in freshwater (S_c = 600) (mean k₆₀₀, diamonds, cm h⁻¹). Error bars associated with k₆₀₀ correspond to spatial heterogeneity in water pCO₂. Water pCO₂ has been measured by the equilibrator technique; air pCO₂, turbulent fluxes (Fₜ, LE, and H), and Uₐ₀ have been measured by the EC technique; and k₆₀₀ has been computed from both technique measurements. All values have been averaged over 10 min.

10 Water pCO₂ in the várzea was always above the observed atmospheric value (387 ± 8 ppmv; Table 1), with an average of 800 ± 75 ppmv and values between 660 ppmv (21 November 2011 at 12:00, measurement number n = 53 in Figure 2) and 1030 ppmv (22 November at 10:30, n = 80) (Figure 2b). High pCO₂ gradients between air and water were therefore observed (415 ± 72 ppmv on average) with a minimum of 300 ppmv. Spatial variations in water pCO₂ within the footprint of EC measurements were weak over the 4 days, with standard deviations ranging from 17 ppmv (20 November, P.M.; Figure 1d) to 72 ppmv (22 November, A.M.; Figure 1g). Significantly higher water pCO₂ values with a mean of 874 ± 70 ppmv (Figure 2b) occurred only on 22 November (p < 0.05, Kruskal-Wallis and Dunn’s multiple comparison tests).
During the whole study, only positive flux values were measured, from 0.05 to 2.2 μmol m⁻² s⁻¹, on 21 November 2011 at 15:00 (n = 63) and 10:10 (n = 42), respectively (Figure 2e). The mean lake CO₂ outgassing flux was 0.59 ± 0.39 μmol m⁻² s⁻¹ (Table 1). The LE heat and H heat fluxes in daytime conditions were generally high, with averages of 198.3 ± 74.1 and 19.1 ± 10.4 W m⁻², respectively (Table 1). Contrary to CO₂ fluxes, significant variations were observed in LE heat and H heat fluxes between 20 November (from n = 14 to n = 41) and 21 November (from n = 42 to n = 76) and in H fluxes between 19 November (from n = 1 to n = 13) and 20 November (Figures 2d and 2e). H fluxes generally correlated with PAR values as for instance on 21 November around noon with values above 40 W m⁻² (Figure 2e). Over the 4 days, \( k_{600} \) evolved from 1.3 to 31.6 cm h⁻¹ with a mean of 12.2 ± 6.7 cm h⁻¹ (Table 1). The associated average wind speed was 3.1 ± 1.7 m s⁻¹. Uncertainties in \( k_{600} \) associated with spatial heterogeneity of water \( pCO_2 \) were below 2.0 cm h⁻¹ on average (Table 1). Finally, significant linear regressions between the gas transfer velocity \( k_{600} \) and the wind speed were obtained only when turbulent sensible heat and latent heat fluxes were low, i.e., below 18 and 180 W m⁻², respectively (Figures 3a and 3b). Unexpected high \( k_{600} \) values were obtained during concomitant low \( U_{10} \) and high heat flux (LE and H) as for instance on 19 and 21 November in the morning (Figures 2a, 2d, 2e, and 2f). Under these conditions, i.e., \( U_{10} \) values below 2.7 m s⁻¹, significant linear regressions between \( k_{600} \) and heat fluxes (sensible heat and latent heat fluxes) were also obtained (Figures 3c and 3d).

### 4. Discussion

Our data set allows computing \( k_{600} \) in an Amazon floodplain lake from concomitant water \( pCO_2 \) and turbulent CO₂ fluxes measured for the first time by equilibrator and EC, respectively. Our \( k_{600} \) values were higher than those previously reported over temperate, boreal, and tropical lakes. Anderson et al. [1999] presented \( k_{600} \) values ranging from 1 to 15 cm h⁻¹ derived from EC measurements over a small woodland lake. Based on EC measurements carried out over a boreal unproductive lake, Jonsson et al. [2008] reported a median \( k_{600} \) value of 7.0 ± 0.6 cm h⁻¹ for an average \( U_{10} \) of 3.9 m s⁻¹. Using floating chamber and EC measurements, Guérin et al. [2007] estimated \( k_{600} \) values averaging 2.9 ± 2.12 cm h⁻¹ over a tropical lake. Over a large Amazonian floodplain lake during the low hydrological phase, Rudorff et al. [2011] computed \( k_{600} \) values averaging 6.2, 12.8, and 11.7 cm h⁻¹ based, respectively, on the parameterization
described by Cole and Caraco [1998], a surface renewal model, and a wind-based model accounting also for diel heating and cooling [MacIntyre et al., 2010]. We confirm here with experimental data that the $k_{600}$ of 2.7 ± 1.0 cm h$^{-1}$ used by Richey et al. [2002] for estimating CO$_2$ outgassing from the Amazonian floodplain is largely underestimated. This latter value came from floating chamber measurements in Amazonian lakes and was consistent with those computed with the wind-based model of Cole and Caraco [1998] deduced from SF$_6$ injection in a temperate lake. Although it is generally hypothesized that chambers overestimate $k_{600}$ by generating artificial turbulence [Vachon et al., 2010], they may also have a significant impact on thermal conditions at the water-air interface and in the case of tropical lakes may inhibit the gas transfer [Guérin et al., 2007].

The $k_{600}$-U$_{10}$ relationship found in Lake Canaçarini was well above the relationships obtained in temperate and tropical lakes by Cole and Caraco [1998] and Guérin et al. [2007] (Figures 3a and 3b). That $k_{600}$ correlated with wind speed only at low heat fluxes confirms the thermal control on $k_{600}$ in addition to wind. In a wind tunnel [Liss et al., 1981] and over the Pacific Ocean [Ward et al., 2004], the gas transfer velocity could be increased by more than 30% under evaporative conditions due to near-surface destabilization. Eugster et al. [2003] also showed in a mid-latitude Swiss lake that convective mixing associated with water heat losses could generate turbulence at the air-water interface, thereby enhancing the gas transfer between the lake and the atmosphere, contrary to stratification associated with water heat losses. MacIntyre et al. [2010] combined EC results from Jonsson et al. [2008] in a Swedish stratified lake and a mechanistic approach to show that $k_{600}$ depended on buoyancy fluxes under nightwind low wind conditions. Rudorff et al. [2011] applied the wind-based parameterization of Cole and Caraco [1998], the surface renewal model, and the wind and buoyancy–flux–based model of MacIntyre et al. [2010] to the thermal conditions of an Amazonian floodplain lake and could quantify theoretically the influence of buoyancy fluxes on $k_{600}$. Here we could compute from data measured directly $k_{600}$ values close to or above 20 cm h$^{-1}$ under very low wind conditions (below 2.7 m s$^{-1}$), concomitant with high sensible heat and latent heat fluxes (Figures 3a and 3b). In these low wind speed conditions, the influence of heat fluxes on $k_{600}$ was unequivocally demonstrated by significant linear regressions obtained with H and LE (Figures 3c and 3d). During our experiment at a wind speed <2.7 m s$^{-1}$, $k_{600}$ increased by 7 cm h$^{-1}$ when H increased by 10 W m$^{-2}$ (Figure 3c) and by 8 cm h$^{-1}$ when LE increased by 100 W m$^{-2}$ (Figure 3d). Several previous studies have hypothesized an enhancement of $k_{600}$ with thermal exchanges, but they could not demonstrate it unequivocally, as their only available proxy for heating and cooling was the water-air temperature gradient [Cole and Caraco, 1998; Guérin et al., 2007; Jonsson et al., 2008; Vachon et al., 2010]. Another advantage of the EC technique is thus to provide a direct measurement of latent heat and sensible heat fluxes that can be correlated with $k_{600}$.

To date, gas transfer coefficients over Amazonian floodplain lakes have been estimated only from chamber measurements or by theoretically applying wind speed–based approaches, surface renewal, and wind/buoyancy flux models for observed meteorological lake conditions [Rudorff et al., 2011]. The present study brings the first $k_{600}$ data over Amazonian lakes obtained by direct equilibrator and EC measurements. To estimate the global Amazon CO$_2$ outgassing flux, Richey et al. [2002] used a $k_{600}$ value five times lower than the average value observed during our measurements (Table 1). Our experimental $k_{600}$ values perfectly match those estimated from wind, surface renewal, or buoyancy/heat flux–based models in another Amazonian floodplain [Rudorff et al., 2011]. As floodplains represent more than 70% of the Amazon flooded area, our results suggest that the regional carbon budget of Amazonian floodplains could be much higher than previously estimated [Richey et al., 2002]. However, one should keep in mind that 70% of the floodplain area is occupied by a flooded forest [Richey et al., 2002], where surface water is protected from wind and solar radiation and where $k_{600}$ might be much lower than in open waters.

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