# A source of methane from upland forests in the Brazilian Amazon

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[1] We sampled air in the canopy layer of undisturbed upland forests during wet and dry seasons at three sites in the Brazilian Amazon region and found that both methane ($CH_4$) and carbon dioxide ($CO_2$) mixing ratios increased at night. Such increases were consistent across sites and seasons. A canopy layer budget model based on measured soil-atmosphere fluxes of $CO_2$ was constructed to estimate ecosystem $CH_4$ emission. We estimate that net $CH_4$ emission in upland forests ranged from 2 to 21 mg $CH_4$ $m^{-2}$ $d^{-1}$. While the origin of this $CH_4$ source is unknown, these ground based measurements are consistent with recent findings based on satellite observations that indicate a large, unidentified source of $CH_4$ in tropical forest regions. **Citation:** Carmo, J. B., M. Keller, J. D. Dias, P. B. Camargo, and P. Crill (2006), A source of methane from upland forests in the Brazilian Amazon, *Geophys. Res. Lett.*, *33*, L04809, doi:10.1029/2005GL025436.

## 1. Introduction

[2] Methane ($CH_4$) is a radiatively active trace gas whose atmospheric budget has been perturbed by human activities. The current global burden of ~4850 Tg $CH_4$ has an atmospheric lifetime of ~8.4 y [*Prather et al.*, 2001]. Chemical reaction with OH is the main sink of $CH_4$ and upland soils are a minor sink. Approximately 600 Tg $CH_4$ $y^{-1}$ are released to the atmosphere [*Prather et al.*, 2001]. Wetlands are the main natural source. The atmospheric burden of $CH_4$ has increased since the industrial revolution [*Etheridge et al.*, 1998]. Over the past two decades the annual growth rate has been highly variable [*Dlugokencky et al.*, 1998, 2003]. The cause of this variability is unknown. Model simulations by *Dlugokencky et al.* [2003] are in reasonable agreement with atmospheric observations in polar latitudes, but agreement is poor in the tropics.

[3] A recent study using the SCIAMACHY instrument aboard the European Space Agency ENVISAT satellite indicated that atmospheric column-averaged $CH_4$ mixing ratios over tropical forest exceeded modeled mixing ratios [*Frankenberg et al.*, 2005] for August through November, 2003. Excess $CH_4$ was obvious over the Amazon Basin. Underestimates in wetland emission inventories may account for part of the discrepancy between the satellite retrievals and modeled estimates [*Melack et al.*, 2004]. Wetlands may not be the only source that has been underestimated. We present data indicating a $CH_4$ source in upland forests based on measurements of ambient $CH_4$ profiles through the canopy layer at three sites across the Amazon Basin.

## 2. Study Areas

[4] Dry and wet season measurements were made at three towers used for micrometeorological studies in upland old growth forest in the Brazilian Amazon in the Brazilian led Large Scale Biosphere-Atmosphere Experiment in Amazonia (LBA) [*Keller et al.*, 2004] (Table 1). The Caxiuanã National Forest site, located near Melgaço, has mean annual precipitation of 2060 mm and a dry period from September through November. The Manaus site in the Cuieiras Reserve has mean annual precipitation of 2250 mm with a dry season from July to September. The Sinop site is located north of that city on a 20 $km^2$ patch of undisturbed forest surrounded by selectively logged forest. Mean annual rainfall is 2037 mm with four dry months from June through September. Annual mean temperatures across sites range from 24.1 to 26.7°C. Studies of ecosystem-atmosphere exchanges of energy, water and $CO_2$ at these site have been described by *Carswell et al.* [2002], *Araújo et al.* [2002], *Vourlitis et al.* [2001, 2002] and *Priante Filho et al.* [2004].

## 3. Methods

[5] We developed a portable profile system for gas measurement and sampling. Four ~6 mm o.d. nylon tubes were mounted on each tower at heights of 5 to 45 m and two tubes were placed at 0.2 and 1 m height about 10 m distant from the tower base. Teflon filters (0.45 μm pore size, Cole Parmer, Vernon Hills, IL, USA) protected each tube inlet. Six levels were sampled at Caxiuanã and Sinop. Only five levels were sampled at Manaus. A valve manifold received six tubes that were flushed continuously at 1 L $min^{-1}$ using separate pumps (UNMP08L, KNF, Trenton, NJ, USA). Sequential selection of each tube outflow to an infra-red gas analyzer (IRGA) (Li-6262, LiCor, Lincoln, NE, USA) allowed us to monitor $CO_2$ mixing ratios corrected for the effects of water vapor in real time. The IRGA was calibrated with secondary standards traceable to NOAA CMDL standards before and after each campaign. Span and zero drifts were less than 1 ppm. The analytical accuracy was traceable to NOAA CMDL standards. The precision expressed as the standard error of the mean for multiple measurements from standards was better than 1 ppm $CO_2$. Air samples were

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**Table 1.** Characteristics of Sample Collections for Three Tower Sites[a]

| Site | Latitude | Longitude | Wet Season | Dry Season | Canopy Height, m | Sampling Heights, m |
|---|---|---|---|---|---|---|
| Caxiuanã | −1.718 | −51.460 | 10–13 May 2005 | 5–8 Nov 2004 | 35 | 0.2, 1, 5, 10,[b] 20,[b] 37[b] |
| Manaus | −2.609 | −60.021 | 12–16 Jan 2004 | 2–5 Aug 2004 | 35 | 0.2, 1, 5, 25, 45 |
| Sinop | −11.413 | −55.325 | 19–22 Apr 2004 | 2–5 Jul 2004 | 29 | 0.2, 1, 5, 10, 20, 32 |

[a]Canopy height refers to the maximum height of the trees in the vicinity of the tower.
[b]During the dry season at Caxiuanã, the top three heights were 40, 25, and 15 m.

collected from each level 3–5 times on several nights and at least once during well-mixed daytime conditions during each campaign for a total of 75 profiles on 19 dates. Air samples were pressurized to 2 atm using a secondary pump (MOA-P101-HJ, GAST Manufacturing, Benton Harbor, MI, USA) into 500 ml electropolished stainless steel canisters closed with stainless steel valves (SS-14DPM-A, Swagelok, Solon, OH, USA) and stored for up to 10 days prior to analysis. Tests of air samples and standards lasting up to 60 days showed no appreciable drift.

[6] Samples were analyzed for $CH_4$ using flame ionization gas chromatography off site [*Varner et al.*, 2003]. An automated analysis routine was used to inject canister samples and $CH_4$ standards traceable to NOAA CMDL standards. Samples and standards passed through Drierite® to remove water vapor prior to analysis. Each canister sample was analyzed at least twice. Analytical accuracy was better than 0.02 ppm $CH_4$ and precision was better than 0.005 ppm expressed as the standard error of the mean for multiple measurements of standards.

[7] We calculated a height weighted average mixing ratio of each gas for the canopy layer (defined as ground level to maximum sampling height) by linear interpolation between levels. We estimated $CH_4$ emission using a canopy layer budget [*Trumbore et al.*, 1990] calculated as

$$\frac{dC}{dt} = P - k(C - C_t) \tag{1}$$

where $C$ is the mixing ratio of the gas in the canopy layer, $P$ is the net production in that layer including the soil-atmosphere flux, $k$ is an exchange coefficient between the canopy layer and the overlying atmosphere and $C_t$ is the mixing ratio of the gas in the overlying atmosphere. By division of equation (1) for $CH_4$ by the similar equation for $CO_2$ we define the following relation

$$\frac{d[CH_4]}{d[CO_2]} = \frac{P_{CH4} - k\left([CH_4] - [CH_4]_t\right)}{P_{CO2} - k\left([CO_2] - [CO_2]_t\right)}. \tag{2}$$

When exchange ($k(C - C_t)$) is small relative to the trace gas emission then

$$\frac{d[CH_4]}{d[CO_2]} \approx \frac{P_{CH4}}{P_{CO2}}. \tag{3}$$

We assume that transport processes for all trace gases between forest canopy layer and the overlying atmosphere are similar [*Oke*, 1987]. The quantity $d[CH_4]/d[CO_2]$ at each site for each season was calculated by orthogonal linear regression of height weighted average gas mixing ratios in the canopy layer using JMP IN software (Version 5.1, SAS Institute, Cary, NC). Orthogonal linear regression requires an estimate of the ratio of the measurement precision of the two variables including analytical and sampling uncertainties [*Tan and Iglewicz*, 1999]. We estimated this ratio by comparison of duplicate profiles.

[8] Soil-atmosphere fluxes of $CO_2$ ($J_{CO2soil}$) were measured at all sites using an open dynamic chamber [*Varner et al.*, 2003] with an IRGA (Li-820, LiCor, Lincoln, NE USA). During each sampling campaign, we measured $CO_2$ fluxes on 16 transects at 8 randomly selected locations per transect (30 m) over a period of about one week for a total of 128 individual flux measurements per campaign. Transects were distributed across the landscape to capture the variation in soils and topography within a ~1 km radius of the towers. Ecosystem $CO_2$ emission was estimated by the ratio ($\rho = 2.4$) of ecosystem flux to soil flux based on ecosystem component respiration studies at the Manaus site [*Chambers et al.*, 2004]. The estimate for ecosystem-atmosphere $CH_4$ flux was then calculated as

$$P_{CH4} \approx \rho J_{CO2soil} \frac{d[CH_4]}{d[CO_2]}. \tag{4}$$

## 4. Results

[9] Figure 1 shows $CO_2$ and $CH_4$ profiles from one day of wet season sampling at Sinop. The height weighted

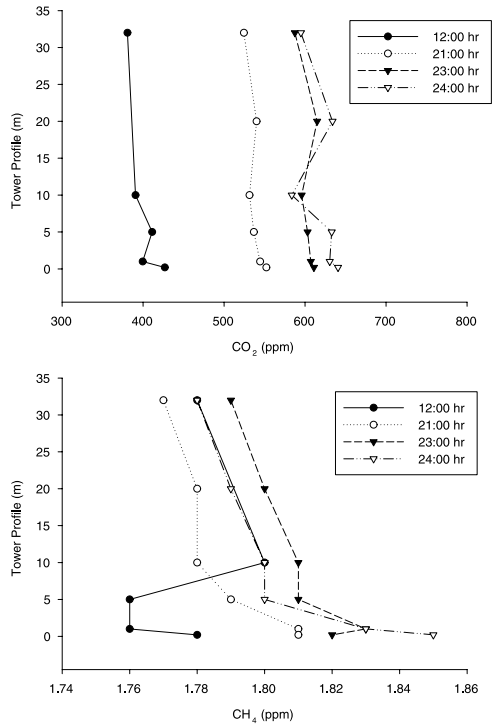


**Figure 1.** Mixing ratios of $CO_2$ and $CH_4$ measured on gas samples from 6 tower levels during daytime and nighttime on 2 July 2004 near Sinop.

**Table 2.** The Ratio $d[CH_4]/d[CO_2]$ Calculated For Six Sampling Periods at Three Amazon Sites and the Corresponding Flux of $CH_4$ Assuming the Estimated Nocturnal $CO_2$ Fluxes at These Sites[a]

| Site | Season | $d[CH_4]/d[CO_2]$ | Soil $CO_2$ Flux (± Std Err.), μmol $m^{-2}$ $s^{-1}$ | $CH_4$ Flux, mg $CH_4$ $m^{-2}$ $d^{-1}$ |
|---|---|---|---|---|
| Caxiuanã | Wet | 0.0010[a] | 6.1 (1.4) | 20.6 |
| Caxiuanã | Dry | 0.0004 | 5.1 (1.5) | 6.9 |
| Manaus | Wet | 0.0002 | 5.4 (1.4) | 3.6 |
| Manaus | Dry | 0.0002 | 5.5 (1.1) | 3.7 |
| Sinop | Wet | 0.0001[a] | 5.8 (1.1) | 2.0 |
| Sinop | Dry | 0.0005[a] | 3.0 (0.5) | 5.1 |

[a]Regressions that are statistically significant ($p < 0.05$).

profile mixing ratios of $CH_4$ and $CO_2$ from all Sinop wet season samplings were significantly correlated ($p < 0.0001$) (Table 2). We found that height-weighted average canopy layer $CH_4$ mixing ratios increased as $CO_2$ mixing ratios increased for all of our sampling campaigns (Figure 2). The average values of $d[CH_4]/d[CO_2]$ for all six sampling campaigns ranged over an order of magnitude (Table 2).

[10] Average soil-atmosphere $CO_2$ fluxes ranged from 3 to 6 μmol $m^{-2}$ $s^{-1}$ for the six sampling campaigns (Table 2). Wet season fluxes exceeded dry season fluxes with the exception of Manaus where fluxes were nearly identical.

[11] Estimated ecosystem $CH_4$ emissions ranged from 2 to 21 mg $CH_4$ $m^{-2}$ $d^{-1}$ (Table 2) with no clear difference between wet and dry seasons. The estimated $CH_4$ emission at Caxiuana was greater than at either Manaus or Sinop which had nearly equal average $CH_4$ emissions.

## 5. Discussion

[12] Nocturnal temperature inversions occur frequently in lowland tropical forests leading to the formation of shallow nocturnal boundary layers [*Garstang and Fitzjarrald*, 1999]. At night, we consistently measured increased mixing ratios of $CO_2$ and $CH_4$ in the canopy layer compared to well-mixed daytime background conditions. Studies from tropical forests have demonstrated that $CO_2$ accumulated in the nocturnal boundary layer [*Trumbore et al.*, 1990; *Culf et al.*, 1999; *Kuck et al.*, 2000]. The consistent correlation that we observed for $CH_4$ with $CO_2$ strongly suggests that there is a widespread source of $CH_4$ in the canopy layer of upland Amazon forest sites.

[13] The Amazon region is well-known for its extensive wetlands that produce large quantities of $CH_4$ [*Bartlett et al.*, 1988; *Melack et al.*, 2004]. We specifically avoided floodplains and lowland sites. None of our sites are subject to inundation and all sites are located at local topographic highs. There is no evidence for a wetland origin of the nocturnal $CH_4$ accumulation measured in this study.

[14] We estimated $CH_4$ emission rates that varied by an order of magnitude across sites and seasons. This range encompassed the variability inherent in natural ecosystem processes as well as random experimental uncertainties and biases in the model used for calculation. While analytical and sampling uncertainties are small (see Methods), our simple model for estimation of $CH_4$ emission ignores horizontal advection as well as exchange between the canopy layer and the air above the canopy layer. These are minor concerns because they should cause a proportional dilution for both $CO_2$ and $CH_4$. Soil $CO_2$ fluxes are quantified to within about 20% (Table 2).

[15] The largest potential bias in our estimate is the adjustment factor (ρ) [*Chambers et al.*, 2004]. The factor ρ is likely to vary among sites, seasons, and years. *Chambers et al.* [2004] note that their average soil flux (3.2 μmol $m^{-2}$ $s^{-1}$) was low compared to other studies in Amazon forests possibly because soils were wet during their measurement year because of heavy precipitation. We measured a soil-atmosphere flux at Manaus of 5.5 μmol $m^{-2}$ $s^{-1}$. If we use this flux and assume that other fluxes measured by *Chambers et al.* [2004] remain constant, the factor ρ decreases to 1.8 thereby reducing $CH_4$ emission estimates by about 30%.

[16] Tropical soils almost always consume atmospheric $CH_4$ [*Keller et al.*, 1986; *Steudler et al.*, 1996]. Tropical forests are the habitat for termites that are known to produce $CH_4$ [*Zimmerman et al.*, 1982; *Fraser et al.*, 1986]. These emissions are difficult to quantify [*Collins and Wood*, 1984] and measurements from tropical forest soils in Cameroon and Borneo indicated that on average soil $CH_4$ consumption dominated over termite produced $CH_4$ [*MacDonald et al.*, 1999]. *Martius et al.* [1993] studied arboreal wood feeding *Nasutitermes* termites in the Amazon. The $CH_4$ emissions from that study were lower than the ecosystem $CH_4$ emission estimated in our study. Other potential $CH_4$ sources include anaerobic decay of waterlogged wood [*Zeikus and Ward*, 1974], poorly drained patches of soil [*Keller et al.*, 2005] and other areas where anaerobic conditions may develop such as thick moss mats on large branches and in the cavities of tank bromeliads.

[17] The dispersed source of $CH_4$ that we identified in upland Amazon forests may partly explain the anomalously high $CH_4$ mixing ratios found in retrievals from SCIAMACHY [*Frankenberg et al.*, 2005]. They indicated a missing source mainly in broad-leaf tropical forests as large

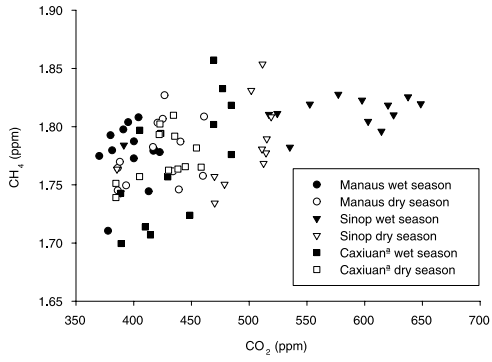


**Figure 2.** Height-averaged canopy layer mixing ratios of $CH_4$ versus $CO_2$ for all profiles.

as 90 Tg $y^{-1}$. Our estimates for $CH_4$ emission ranged from 2 to 21 mg $CH_4$ $m^{-2}$ $d^{-1}$. We recognize that our results are sparse and that the upper estimate would fall by a factor of three if we excluded the Caxiunã wet season campaign. Multiplying these source estimates by an upland forest area for the Amazon region of $5 \times 10^6$ $km^2$, resulted in an annual $CH_4$ flux of between 4 and 38 Tg $y^{-1}$. Even the lower estimate of this $CH_4$ emission represents nearly 1% of global $CH_4$ emissions.

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