

Net fluxes of CO₂ in Amazônia derived from aircraft observations

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ABSTRACT

A conceptual framework is developed using atmospheric measurements from aircraft to determine fluxes of CO₂ from a continental land area. The concept is applied to measurements of CO₂, O₃, and CO concentrations from the Atmospheric Boundary Layer Experiment, (ABLE-2B, April-May 1987), to estimate fluxes of CO₂ for central and eastern Amazônia late in the wet season of 1987. We observed that column amounts of CO₂ decreased during the day over the Amazon Basin at the average rate of $-6.3 \pm 1 \mu\text{mole m}^{-2} \text{ s}^{-1}$, corresponding to an uptake flux somewhat smaller than the daytime uptake ($-10.2 \mu\text{mole m}^{-2} \text{ s}^{-1}$) at a flux tower in the study area. The estimated net flux of CO₂, integrated over 24 hours, was $-0.03 \pm 0.2 \mu\text{mole m}^{-2} \text{ s}^{-1}$, small compared to model results for seasonal uptake of CO₂ or to long-term sinks attributed to the region. The results indicate that the carbon budget of Amazônia was close to balance in April 1987. We argue that, at the scale of the whole Basin, seasonal hydrological factors modulate respiration on the forest floor and in wetlands, offsetting growth of forest trees, and thus it is essential to consider landscape mosaics and seasonal modulation of decomposition processes when assessing CO₂ fluxes at continental scale.

Introduction

Analysis of the budgets for atmospheric CO₂, O₂, and ¹³CO₂ indicate that forests have taken up > 2 PgC/yr of fossil fuel on average over the last 20 years [Battle *et al.*, 2000]. Inverse models point towards a sink in Northern temperate forests [Tans *et al.* 1990; Fan *et al.*, 1998; Ciais *et al.*, 1995; Rayner *et al.*, 1999; Bousquet *et al.*, 2000], but forest inventories [e.g. Houghton *et al.*, 1999] suggest much smaller uptake. Alternatively, Phillips *et al.* [1998] and Malhi and Grace [2000] propose that mature Amazonian forests represent a major sink for global CO₂, up to 2 Pg C/yr, based on data from ecological plots and eddy correlation flux towers. They cite rising CO₂ concentrations as a possible stimulus for carbon uptake in tropical forests.

Conflicting claims for the loci of carbon sinks have been difficult to test due to the lack observations over the continents. Most atmospheric CO₂ data are obtained at surface sites in remote oceanic locations, selected deliberately to minimize the influence of continental sources. Measurements close to sources and sinks are subject to large diurnal and spatial variations, due to daily alternation of uptake and release by vegetation and to variation of surface fluxes over the landscape mosaic. Hence very few observations exist over the continents. Thus *a priori* specification of flux patterns over the globe play a critical role in inverse models [Kaminski and Heimann, 2001; Rayner *et al.*, 1999], and results are influenced by the prevailing consensus favors uptake of CO₂ by mature tropical forests that significantly offsets losses due to deforestation. Tests of this view using large-scale atmospheric data over land have not been attempted.

The present paper introduces a new conceptual framework to use aircraft data to obtain direct estimates for regional fluxes of CO₂ over a continent. We use mass balance calculations for the atmospheric column for each hour of the day, and account for CO₂ from combustion and from long-range meridional transport using data for CO. The concept is applied to an historical data set in Amazonia, to derive values for the rate of daytime uptake by the mosaic of surface vegetation and to estimate regional net 24-hour flux of CO₂. The work extends the analysis of column integrals presented for the earlier ABLE-2A in 1985 [Wofsy *et al.*, 1988]. The results suggest that the net flux for CO₂ in Amazonia was ~0 in April 1987 [Chou, 1999].

Observations

The Amazon Boundary Layer Experiment (ABLE-2B, April-May 1987) was conducted in the Brazilian Amazon in April and May 1987, months 6 and 7 of the 9-month wet season. Instruments on board the NASA Electra aircraft measured CO₂, O₃, CO, NO, aerosols, and meteorological parameters for 21 missions of four types [Harriss *et al.*, 1990]: *Surveys* flew east from Manaus to Belém and back (see Figure 1). *Source* missions intensively sampled the lower atmosphere (0 – 3 km) over selected vegetation, e.g. forest or wetland, principally north of Manaus. *Convective transport* flights covered a wider altitude range to examine the influence of convection. The *flux* mission obtained eddy-correlation fluxes on level runs in the Planetary Boundary Layer (PBL). A total of 85 vertical profiles were obtained, with dense, repeated coverage in central equatorial Amazonia (1 – 4 S) and two cross-sections upwind from the main sampling area to the Atlantic coast (Figure 1).

Data for CO₂ were obtained on 15 missions spanning hours from 0700 to 1700 local time (GMT – 4 hours) at altitudes from 0.15 to ~6 km. Included were seven *source* missions (one principally over wetlands), four *transport* missions (“volume” flight pattern), three *survey* missions, and the *flux* mission [see Table 1 and Chou, 1999]. Most data were acquired between 0.15 and 3.1 km.

Concentrations of CO₂ were measured using a BINOS non-dispersed infrared analyzer. An upstream Teflon-diaphragm pump drew air from the inlet, into a wet trap at 0° C, to set a constant dew point, then through a pressure-control solenoid valve (MKS capacitance manometer and MKS 250B pressure controller) that maintained the pressure at 740 torr in the wet trap and associated plumbing. A small portion of this flow was drawn into the sample cell by a pump downstream of the analyzer. Constant pressure was maintained in the cell using a second manometer and pressure controller, located downstream of the analyzer and upstream of the pump. Reference gas with near-ambient concentration of CO₂ in dry air was passed through the reference cell, maintained at the same pressure as the sample cell. The response time for the instrument to a change in concentration at the inlet was 1-2 s, depending on altitude, corresponding to resolution ~200m in the horizontal and ~10m in the vertical.

Calibrations were carried out frequently in flight by flowing standard gases (CO₂ in air, Standard Reference Materials obtained from the National Bureau of Standards) into the inlet, upstream of the wet trap. Calibrations were routinely performed at the beginning, midpoint, and end of each vertical profile to insure unbiased results with respect to altitude. Instrument drift was generally less than ±0.1 ppm during a flight. Calibration gases, stated to be accurate to ±2 ppm, bracketed observed concentrations in the atmosphere. One set of standards was used throughout, insuring uniform measurements during the 24 days of observations.

Corrections were applied to account for BINOS sensitivity to aircraft attitude and acceleration by fitting data from calibrations to a 2nd-order polynomial function of the instantaneous acceleration vector, obtained from a tri-axial linear accelerometer mounted on the instrument. Corrected instrument response had residual sensitivity to rotational accelerations in turbulence and vertical spirals, amounting to a few tenths of 1 ppm, occasionally (in sharp turns) by as much as ±0.5 ppm.

Concentrations of O₃ were measured using ethylene C₂H₄ chemiluminescence [Gregory *et al.*, 1990], and concentrations of CO were measured by the Differential Absorption CO Measurement (DACOM) system, a fast-response tunable diode laser spectrometer [Harriss *et al.*, 1990b]. The trace-gas instruments did not report accurate concentrations during intervals when eddy flux measurements were in progress, since proper calibrations were not possible at the large flow rates required for eddy fluxes. Data for other ABLE2B observations, including CO, O₃, and CO₂, were averaged into 10s intervals for archiving and merging into a unified data set.

Measurements for CO₂ and O₃ eddy covariance fluxes at 41 m, and concentrations at 0.02, 3, 6, 12, 27, 36, 41 m, were obtained at a tower in Ducke reserve on the outskirts of Manaus [Bakwin *et al.*, 1990; Fan *et al.*, 1990]. Results were reported for ~8 days for non-precipitating conditions during the experiment. The fetch for this tower was predominantly upland forest with no large watercourses or wetland areas.

Table 1. Electra Aircraft Missions During ABLE 2B

FLT	Time (local)	Day of 1987	Mean Lat (°S)	Mean Lon (°W)	Mission Type [Browell <i>et al.</i> , 1990]	Altitude Range (km)
6	13-17	105	2.4	60	Transport-double wall	0.3-4.7
7	10-15	107	2.7	60	Transport-volume	0.2-4.6
8	10-14	109	3.8	60	Source	0.2-4.6
9	12-17	110	1.8	59	Source	0.2-4.6
11	8-10	113	2.1	53	Manaus-Belèm survey	0.2-3.0
12	7-11	114	NA	51	Belem-Santarèm survey	0.2-3.0
13	14-17	114	3.0	58	Santarèm-Manaus survey	0.2-3.0
14	7-10	116	2.5	60	Source	0.2-3.0
15	8-9	118	1.2	59	Source	0.2-3.0
16	8-9	119	2.7	59	Source-forest	1.6-4.3
18	11-16	122	3.0	58	Source-wetlands	0.2-4.3
19	11-13	124	2.8	60	Flux	0.8-3.0
20	9-14	126	2.5	60	Transport-volume	0.2-3.7
21	9	127	2.4	60	Source	0.2-4.4
22	11-16	128	2.4	60	Transport-volume	0.2-4.6

Figure 2 shows typical data, from Flt. 18 over a mix of flooded and upland areas, Flt. 14 over forests northwest of Manaus, and Flt. 12 in eastern Amazônia between Santarèm and Belèm. Combustion plumes were encountered in Flt. 14 (note spikes in CO at 7.9, 9.4, and 9.7 hrs in Fig. 2a), possibly from Manaus. Weaker emissions from biomass fires were seen on many flights. Note the similarity between CO₂ data from the eastern and central parts of the Basin. Figure 2b shows individual profiles, illustrating the reproducibility of the measurements (typically a few tenths of 1 ppm, compare ascent and descent above the PBL). Elevated CO₂ is seen at low altitudes in the morning, as CO₂ from nighttime respiration is mixed upwards into the growing PBL. Low CO₂ concentrations just above the morning PBL (0.6-1.2 km) represent the relict PBL from the previous afternoon, with low CO₂ (due to photosynthesis) preserved by lack of vertical mixing during the night. Differences between CO₂ in the relict PBL and air aloft (≥ 3 km) were much smaller over Amazônia (3-5 ppm) than over North America in summer [Gerbig *et al.* 2001] (10 – 20 ppm or more). Relatively small drawdowns were observed consistently in the afternoon in Amazônia, as seen, for example, on Flt. 18 (Fig. 2b, *lower panel*).

Figures 3a-3c show mean vertical profiles of CO₂, CO and O₃ block averaged by hour and altitude (200 m bins) over the 24 days/15 flights; tower data block-averaged by height and hour of the day were appended at the bottom. The atmosphere steadily loses CO₂ during the day due to photosynthetic uptake, although midday CO₂ concentrations remain higher in the lower atmosphere than aloft. Only late in the afternoon were PBL values depleted, by 1 – 2 ppm, compared to air higher up (Fig. 3a). The data for 1400 Local Time are anomalous above ~1.5 km, reflecting unusual CO, O₃ and CO₂ concentrations on a single flight (#7) which we judge to be

unrepresentative. Apart from these data, the flights were remarkably consistent despite fine-scale variance.

The CO₂ profile averaged over the day shows a weak minimum between 1 and 2 km, levels where exchange with the surface is active only in the afternoon when concentrations are low. There is a corresponding enhancement of daily mean CO₂ at 500m and below (Fig. 3a). This contrast ("rectification") arises from co-varying responses to diurnal forcing of photosynthesis and of growth of the PBL [Denning *et al.*, 1995, 1999]. The magnitude (~ 1 ppm) and vertical extent (~ 2 km) was smaller over Amazônia at this time compared to some model results [Denning *et al.*, 1995]. The density-weighted mean concentration for the whole profile was very close to the value at the top of the profile, a sharp contrast with results from mid-latitudes [Gerbig *et al.*, 2001], as discussed in detail below.

The profiles in Fig. 3a provide a conservative estimate of the errors in defining the mean profiles, omitting data from 1400 hrs. Each altitude is treated independently, hence the variance about a smoothed trend line versus altitude, about ± 0.1 ppm, is a measure of the error in the mean profile.

Concentrations of CO were generally highest near the ground, and typically increased during the day at low altitudes (Figs. 3b, 3d), due to inputs at the surface. Concentrations of O₃ were ~20 ppb at 2 km, declined to 10 ppb at the canopy, and vanished at the ground, reflecting strong uptake by vegetation and weak photochemistry [Fan *et al.*, 1990; Jacob and Wofsy, 1990]. Concentrations of O₃ were generally higher above the canopy in the morning vs. the afternoon, reflecting maximum uptake (regulated by stomatal opening) by vegetation during daytime.

Framework for analysis of aircraft measurements

Basic concept

Air enters the Amazon Basin with the Trade Winds from the east (Atlantic Ocean) and flows towards a major region of convergence in western Amazônia. The observed winds were almost due easterly from ~1 to 10 km throughout the equatorial zone, with maximum speed of 10 m s⁻¹ at 2 km (see Fig. 6 below). Our measurements were made upwind of the main convergence zone, but downwind of moist equatorial forest stretching all the way to the ocean [Santos, 1987]. Rainfall was 200.3 mm during the study [Garstang *et al.*, 1990], ample, but somewhat lower than the long-term mean (~330 mm). Despite significant rainfall, Central and Eastern Amazônia experienced net divergent flow [Greco *et al.*, 1990], and thus there was a small net subsidence (~200 m/day) over the Basin. Convection was confined to small, locally occurring storms on 42% of the study days, with moderate-scale, early-morning systems ("basin occurring systems") on 30% of the days. Thus a typical PBL developed over the Basin in response to solar heating during the day for >70% of the study days [Greco *et al.*, 1990].

Our concept is to derive the mass budget for atmospheric CO₂ and O₃ for the continental region upwind of our operational area, from the ground to fixed height *h*. We adopt a simple conceptual model to illustrate the framework for using aircraft data

to measure continental-scale fluxes. Ideally we would use a high-resolution mesoscale model incorporating detailed representation of actual convective transports, but this is not possible for an historical data set. We envision air flowing into the region from the east, with transit time from the sea of 3 – 5 days. The lower atmosphere accumulates the influence of surface sources and sinks due to vertical mixing in the PBL and associated Convective Cloud Layer (CCL), slowly exchanging with air from aloft by subsidence/divergence and intermittent deep convection.

Exchange with the surface has the strongest influence for altitudes that interact daily with the ground, i.e. the PBL and CBL at their maximum daily height, h . Thus the chemical composition of air below h is strongly influenced by surface fluxes, averaged over a diurnal cycle; air above h reflects the large-scale circulation. Thus the fixed value for h is selected to include all altitudes in daily contact with the surface, on days with well-defined growth and decay of the PBL. We used meteorological and chemical measurements to select h , and we consider two bounding values (2500 m and 3300 m) to insure that results are not affected by this selection.

The concept is expressed mathematically by vertically integrating the mass continuity equation for tracer in a column with unit area and height h , moving with the mean flow,

$$n_b \frac{\partial q_{ib}}{\partial t} + \frac{n_h \times (q_{ib} - q_{ih})}{\tau_{\text{exch}}} = \left(\frac{S_i}{h} + [P_i - L_i] \right) = \underline{S}_i. \quad \text{Eq. (1a)}$$

Here S_i denotes surface flux of the two principal species for this study, CO_2 or O_3 (moles $\text{m}^{-2} \text{s}^{-1}$), $[P_i - L_i]$ is the mean net chemical tendency for altitudes $0 - h$ (moles $\text{m}^{-3} \text{s}^{-1}$), q_{ib} is the mean mole fraction of tracer i between 0 and h ($q_{ib} = \frac{1}{n_b} \int_0^h q_i n dz$), n_b

is the mean atmospheric number density (moles m^{-3}) from 0 to h , n_h is the atmospheric number density at h ($n_{z=h}$), q_{ih} is the mole fraction of tracer at h , and τ_{exch} is the mean time to replace air from 0 to h with air from above h . Time t is measured from the entry of air onto the continent. The "integrated net source", \underline{S}_i , combines the effects of net surface flux and chemical reactions on the mean atmospheric composition for altitudes 0 to h . Note that $z=0$ refers to the top of the forest canopy, since our measurements do not resolve CO_2 sources or concentration gradients in the forest.

Equation (1a) can be used to determine *daytime hourly* values of \underline{S}_i from hourly data for q_{ib} and q_{ih} , provided that entrainment of air from aloft exchanges only a small fraction of the air from 0 to h over a period of 8 hours, which insures that

$n_b \frac{\partial q_{ib}}{\partial t} \approx \left(\frac{S_i}{h} + [P_i - L_i] \right)$. This is usually an excellent approximation for ABLE-2B data, as indicated by mean subsidence velocities and basin-wide convective mass fluxes [Greco *et al.*, 1990] which gave replacement times for the column between 0 and h (τ_{exch}) between 3 and 5 days.

Equation (1a) can be integrated and averaged over 24 hours $\left(\frac{1}{T} \int_0^T \{ \dots \} dt \right)$,

denoted by $\langle \dots \rangle$, $T = 24$ hours) to obtain

$$n_b \frac{q_{ib}(T) - q_{ih}(0)}{T} + \frac{n_h \langle (q_b - q_h) \rangle}{\tau_{\text{exch}}} = \langle \underline{S}_i \rangle. \quad \text{Eq. (1b)}$$

The lower atmosphere in central Amazônia has cycled through 4 – 5 diurnal cycles, sufficient to approach a quasi-periodic state where $(q_{ib}(T) - q_{ih}(0))/T$ is smaller than other terms in Eq. (1b). Thus *24-hour mean net exchange*, $\langle \underline{S}_i \rangle$, can be estimated,

$$\langle (q_{ib} - q_{ih}) \rangle \frac{n_h}{\tau_{\text{exch}}} \approx \langle \underline{S}_i \rangle. \quad \text{Eq. (1b')}$$

The derivation allows for entrainment of air from aloft at the mean rate h/τ_{exch} , and assumes that our tracers, CO, CO₂ and O₃, are well mixed over the ocean ($q_{ib} = q_{ih}$), as observed in ABLE-2A [Wofsy *et al.*, 1988]. Equation (1b') implies a *similarity* relationship for two tracers,

$$\boxed{\frac{\langle q_{1b} - q_{1h} \rangle}{\langle q_{2b} - q_{2h} \rangle} = \left(\frac{\langle \underline{S}_1 \rangle}{\langle \underline{S}_2 \rangle} \right)}, \quad \text{Eq. (1c)}$$

i.e. the *ratio of vertically integrated net sources for two tracers, averaged over 24 hours, can be derived from the ratio of the differences in concentrations between altitude h (q_h) and the mean from 0 to fixed height h (q_b), $\langle q_{ib} - q_{ih} \rangle$.*

Requirements for similarity (Eq. 1c) are discussed below. Similarity arises from the mass balance of the atmospheric column to fixed height h , where there is close spatial and temporal covariance of the S_i for two tracers (sources and sinks are similarly distributed within the fetch). Eq. 1c holds for many realistic transport scenarios as well as the simple illustrative analysis adopted below, if the covariance conditions are satisfied.

In ABLE-2B we obtained data for daytime, from the interval 7-8 hours to 16-17 hours local time, with a few after 17 hours. Our data cover the daily turning points of the column concentrations of CO₂, observed by Wofsy *et al.*, [1988] from aircraft in ABLE-2A (when nighttime data were obtained) at ~ 8 hrs (daily maximum) and between 16 and 17 hours (minimum). The daily turning points from ABLE-2A data were consistent with times for reversal of CO₂ fluxes over the forest, from source to sink in the morning and conversely in the late afternoon (see Figure 3g, from Fan *et al.*, 1990), in ABLE-2B. Flux reversals occurred when photosynthesis rates just balanced respiration from the soil, about 1 hour after sunrise and 1-2 hours before sunset. Thus *our daytime data are sufficient to derive the 24-hour mean values for q_{ib} required for Eqs. (1b-1c), provided that the system satisfies the quasi-periodic condition*, since increases at night will, on average, reverse the daytime drawdown. Close correspondence of data from the cross-basin flights with data from central Amazônia supports application of the quasi-periodic condition (see Fig. 1, *inset*).

Our analysis of column budgets using Eq. (1a-c) differs from the conventional approach that follows concentrations in the Convective Boundary Layer during growth and decay over the day ("CBL method"). In particular, the value of q_b in (1) is not affected by processes that rearrange concentration gradients between the ground and the fixed level h , e.g. by PBL mixing into the residual layer during the morning.

Similarity criteria, selection of h, and accounting for advection and combustion

Since CO₂ is inert in the atmosphere, but O₃ is not, similarity requires that the chemical tendency for O₃ ([P – L]) be small compared to the surface flux S/h. Fluxes of O₃ to the forest were -3.8 and -0.37 nmol m⁻²s⁻¹ in the day and night, respectively [Fig. 3g, *Fan et al.*, 1990], giving $\langle \underline{S} \rangle = -6.8 \times 10^{-13}$ moles m⁻³s⁻¹ for h=3300 (– 8.4 × 10⁻¹³ moles m⁻³s⁻¹ for h=2500m). Model results indicated that the chemical tendency for O₃ averaged 2 × 10⁻¹⁴ moles m⁻³s⁻¹ [*Jacob and Wofsy*, 1990], less than 3% of the deposition flux, because concentrations of NO were very low.

During the day, forests take up CO₂ and O₃ at rates regulated by stomatal conductance. At night, forests emit CO₂ and cease to take up O₃. Rivers and flooded lands are like forests at night: they emit CO₂ [*Richey et al.*, 1990] but exchange little O₃ due to its low solubility.

By comparing daytime changes in column-mean amounts of CO₂ and O₃ with fluxes observed at the Manaus tower, we can test the postulates of the similarity analysis: covariance of CO₂ and O₃ in the lower atmosphere, and surface control of changes in column amounts. Diurnal patterns for surface fluxes at the tower were roughly similar, although uptake of O₃ was apparently more strongly limited by partial closure of the stomates at midday [Fig. 3g, *upper*; *Fan et al.*, 1990] than was CO₂. Column mean concentrations declined for both gases during the day (Fig. 3g, *lower*), following surface fluxes with a delay of 1-2 hr, consistent with the time scale for turnover of the PBL. In fact, concentrations of O₃ recovered somewhat around 2 pm, apparently reflecting midday inhibition of uptake.

This comparison supports the view that changes in column-mean (from 0 to h) concentrations for both gases are driven by surface exchange, with broadly similar diurnal behavior. This analysis is only suggestive, of course, because of differences in scale for tower and aircraft measurements. Nevertheless the available evidence supports the use of Eq. (1c) to derive approximate values for 24-hour net exchange for CO₂, based on observed changes in concentrations of CO₂ and O₃ between 0 and h.

The height of the fixed level h (Figure 4a) was selected by examining profiles for CO, O₃, and H₂O for each flight to estimate the altitudes influenced by surface exchange, representing the height of the PBL plus the layer influenced by detrainment from the tops of fair-weather clouds (Convective Cloud Layer, or CCL). The maximum at 1400 local time, 3.1 km, lay at the base of the trade-wind inversion. Analysis of the diurnal variations of CO₂ concentrations gave a slightly lower value for the maximum height in daily contact with the surface, 2.5 km. We used both values to analyze data from ABLE-2B.

Fine-scale variations of CO₂ were consistently correlated with concentrations of CO, especially in distinct concentration spikes (e.g. Fig. 2, Flt. 14). Some of this covariance was associated with combustion sources within the study area, but some arose from large-scale transport (e.g. inter-hemispheric gradients; Fig. 3f). Concentrations of both CO₂ and CO at 3 km and above were bracketed by concurrent values at Samoa (SMO, 14 S) and Mauna Loa (MLO, 19 N), in the mid-Pacific. Changes in CO₂ and CO from W to E across the Amazon Basin closely approximated

an mixing line with end-members at SMO and MLO (Figs. 3 a,b,f). These data are consistent with the observation [Boering *et al.*, 1994; Andrews *et al.*, 1999] that concentrations of CO₂ in the upper tropical troposphere may be accurately predicted for any month by averaging data from SMO and MLO, with a delay of 2 months at ~16 km. (*Note:* Measurements of CO concentrations at MLO and SMO do not exist for 1987. Since seasonal cycles and latitude gradients for CO are reproducible from year to year, we used CO data for 1990, the first available year.)

Our concept requires that we distinguish changes in CO₂ due to forest metabolism from changes due to combustion, or from variable admixtures of Northern and Southern hemisphere air advected into the study area. We observed that combustion and large-scale mixing produced similar correlations, 0.04 – 0.1 ppm CO₂/ppb CO [cf. Andreae *et al.*, 1988]. We therefore developed a statistical relationship between CO and CO₂ at each altitude to remove the influence of these processes on q_b and q_h . We compared the result to simple conditional sampling that removed high CO values, as described below.

A key assumption in Eq. (1c) is that q_h does not change as air transits across the basin from the Atlantic, despite the influence of deep convection. The Cross-Basin survey data supported this view, once the effects of large-scale mixing were removed. Thus it appears that air entrained into the PBL during transit may be assumed to have concentration q_h . Another assumption is that air leaving the column has the 24-hour column mean concentration, q_b . If convection occurred preferentially at times of day when concentrations of CO₂ in the PBL were quite different than the 24-hour mean value, the transport through level h could have a bias. Fortunately, the distribution of convective rainfall over the day was almost unbiased relative to the diurnal cycle of CO₂. About 45% of rainfall in the Basin occurred between midnight and 1300 LT, when concentrations CO₂ in the lower atmosphere exceed the 24-hour mean, with 55% at times when CO₂ was depleted by photosynthesis in the afternoon [Fig. 4b, Greco *et al.*, 1990]. Hence deep convection transported air aloft with mean CO₂ very similar to the 24-hr mean, biased at most 10% towards lower concentrations. The observed precipitation was notably less biased towards the afternoon hours than predicted by some models [e.g. Peylin *et al.*, 1999].

Linear Models Representing the Aircraft Data Set

Our goal in analyzing these data is to obtain mean profiles representing all 15 flights throughout the Basin for 24 days, averaging out variations associated with weather, particular locations, etc. We also wish to use the CO tracer to remove the influence of combustion and of long-range advection (inter-hemispheric exchange). We adopted several alternative approaches to this averaging problem, to insure that the treatment did not introduce spurious results.

First we took the simplest approach to define mean profiles, using the block-average of *all data* by hour and by altitude (200m intervals), then computed column-mean concentrations q_b and concentrations at h (q_h) for each hour of the day. This procedure does not account for combustion inputs or meridional advection. Next, we

used *conditional sampling*. We removed O₃ and CO₂ data associated with CO values higher than 90 ppb, and block-averaged the remaining data (3400 of 9400 observations) as before. This approach insures that artifacts due to combustion inputs and large-scale advection are small, but slight biases could remain. These two methods were used to check that our procedures to account for systematic covariance of CO and CO₂ did not introduce artifacts. In fact the results were very close (within 0.1 - 0.2 ppm) to those obtained using the more complete treatment described below.

Our third method derived linear functions to represent data for CO₂ and O₃ in each 200-m altitude band, using three predictors, *hour* of the day, *altitude*, and *CO*, following *Potosnak et al.*, [1999]. We treated hour and altitude as discrete *factors* and the concentration of CO as a continuous linear predictor, with one coefficient for all hours at each altitude. The equations for this approach ("Linear Model"), for the j^{th} 200 m altitude band, are:

$$[\text{CO}_2]_j = a_{j0} + a_{j1} [\text{CO}]_j + \sum_i a_{ji} \delta_{ti} , \quad \text{Eq. (2a)}$$

$$[\text{O}_3]_j = b_{j0} + b_{j1} [\text{CO}]_j + \sum_i b_{ji} \delta_{ti} . \quad \text{Eq. (2b)}$$

Here $[\text{CO}_2]_j$ and $[\text{CO}]_j$ denote observed concentrations in 10s intervals falling in the j^{th} altitude band at hour t (truncated to the nearest hour). The terms $\sum_i a_{ji} \delta_{ti}$ or $\sum_i b_{ji} \delta_{ti}$ represent mean concentrations at each altitude in the (i^{th}) hour (7 to 17, local time), a_{ji} or b_{ji} . Values of coefficients {a} and {b} were optimized using generalized regression for eighteen 200-m altitude bands (Tables 2 and 3), with 10 time-of-day factors for each altitude (the mean for 7 h at altitude j is absorbed into a_{j0} or b_{j0}).

Equation (2) with optimized coefficients provides mean CO₂ and O₃ at each altitude for each hour, and the mean dependence on CO at each altitude. We then generate synthetic CO₂ profiles, in which the effects of combustion sources and advected CO₂ are removed by setting CO to its background concentration, as *Potosnak et al.* [1999] did for data from Harvard Forest.

The Linear Model functions generally fitted the observations well considering the composite treatment of data for a month (see Table 2, and fitted points for typical flights in Fig. 2). If we allow for residual variance from the CO₂ instrument, 1-2 ppm, the Linear Model accounted for more than 60% of observed atmospheric variance up to about 1100 m. Above ~1300 m atmospheric variance is comparable to instrumental variance and it is harder to judge the fidelity of the fits. The coefficients for CO₂ dependence on CO were consistent throughout the lower part of the profile (200-1700 m), with overall mean value 0.078 ± 0.04 (1σ) ppm/ppb. This value lies within the range observed for biomass burning [*Andreae et al.*, 1988] and large-scale mixing (0.04 – 0.1).

Residual variance for O₃ increased with altitude as concentrations increased, in contrast to the results for CO₂. Coefficients for CO in the O₃ fits were variable because the dependence of O₃ on CO was weak (Table 3). Ozone varied less than CO₂ with time-of-day but more from flight-to-flight. To test flight-to-flight variance affected derived values of q_b and q_h for ozone, we constructed another linear model by adding a factor variable for each of the 15 flights. The model produced values of $(\text{O}_{3b} - \text{O}_{3h})$ very similar to results both from the Linear Model and from conditional sampling, and we concluded that variability in background O₃ did not affect the analysis.

Results: Budget of CO₂ over Amazônia

Figures 5a-5d show profiles for CO₂ and O₃ from the Linear Model, with influences of combustion and large-scale exchange removed by setting the concentration of CO in Eq. (2) to the estimated CO background concentration, chosen as the 20th percentile for all flights (84 ppb) following *Potosnak et al.* [1999]. Data from the tower were appended at the bottom. Results were insensitive to the choice of percentile corresponding to the background value: the 10th and 30th percentiles gave 80.3 ppb and 87.7 ppb, respectively, corresponding to ± 0.3 ppm CO₂ or less.

The Linear Model functions provide smooth vertical profiles for each hour, and smooth diurnal variations for each altitude (excepting 1400 hr; see Figs. 4a, 4b). Since each altitude and hour was treated independently, the smooth behavior provides support for the validity of the data averaging. Mean hourly and daily values of q_b and q_h are shown in Figures 5c- 5f, derived from profiles in Figures 5a and 5b by averaging from 0-2500m or 0-3300m. Tower data for 41m were adopted for 0–75m, and values of q_b and q_h at 1400 LT were replaced with the mean of 1300 and 1500 LT as noted above. It appears that CO₂ was added to the column at 1700 hr, consistent with the flux data from the Manaus tower (Fig. 3g) and data from ABLE-2A (Wofsy et al., 1988), but data are too few to give reliable values for q_b at 1700 LT.

If we take the deviation from a smooth curve as an indicator of residual random errors, error for CO₂ concentrations at each altitude for each hour are better than ± 1 ppm. Uncertainties in mean column concentrations, estimated from the standard error of the value at 1200 LT for a linear fit vs. time in Figs. 4 c-f, were less than ± 0.3 ppm. Corresponding values for O₃ were ± 2 ppb and ± 0.5 ppb, respectively.

Table 2. CO₂ Linear Model

Alt. (m)	Mean	S _r	R ²	a _{j1}
100	349.488	15.1	0.74	0.1891
300	350.704	21.8	0.65	0.0875
500	349.282	17.0	0.51	0.0902
700	348.475	7.03	0.45	0.0701
900	347.315	5.05	0.47	0.0823
1100	346.738	3.04	0.41	0.0667
1300	346.663	2.45	0.33	0.0816
1500	347.107	2.54	0.22	0.0692
1700	347.291	2.14	0.36	0.0871
1900	347.360	1.40	0.24	0.0484
2100	347.340	1.24	0.36	0.0468
2300	347.391	0.98	0.33	0.0371
2500	347.449	1.25	0.23	0.0516
2700	347.190	1.11	0.36	0.0428
2900	347.700	1.46	0.46	0.0732
3100	348.334	1.26	0.34	0.0171
3300	347.325	2.13	0.46	0.1123
3500	347.350	2.83	0.69	0.1569
MEAN	347.806	5.00	0.42	0.078

Mean=24 hr. average (ppm).

Table 3. O₃ Linear Model

Alt. (m)	Mean	S _r	R ²	b _{j1}
100	9.564	5.74	0.50	0.1378
300	8.246	6.06	0.22	-0.0057
500	9.446	4.86	0.15	-0.0372
700	12.108	8.44	0.41	0.0466
900	11.733	6.02	0.17	0.0359
1100	13.2465	6.93	0.17	0.0725
1300	14.3656	6.35	0.33	0.0589
1500	15.1017	5.77	0.36	0.0776
1700	15.7222	5.73	0.25	0.0672
1900	17.0503	4.98	0.24	0.0754
2100	16.7096	9.73	0.16	0.0645
2300	17.1151	11.8	0.20	0.0901
2500	17.8334	10.3	0.25	0.0505
2700	18.5788	7.71	0.18	-0.0720
2900	18.9267	11.7	0.39	-0.2068
3100	19.1500	16.1	0.37	-0.2535
3300	19.0895	18.8	0.53	-0.2697
3500	19.3832	17.9	0.56	-0.3299
MEAN	15.187	9.1	0.30	-0.022

S_r, square root of the residual variance (ppm for CO₂, ppb for O₃).

Table 4 gives values for $\langle(q_b - q_h)\rangle$ averaged over daytime (i.e. between the turning points, 0800 – 1700, denoted 8 – 16 hr in Fig. 5), for three treatments: "all data", block averaged by altitude (no linear model or conditional sampling); *Linear Model*, Eq. (2) with no CO adjustment (CO set to the block-averaged mean), presented to show the influence of the CO adjustment; and "*Linear Model, CO_{bkgd}*", Eq. (2) with CO set to 84 ppb. Results for "All Data" and *Linear Model* with CO_{obs} are similar for both CO₂ and O₃ in most cases, indicating fidelity of the linear functions. The *conditional sampling* results were very close to the *Linear Model* with CO_{bkgd}. In almost all cases, using observed CO gives a more positive value for Δq for both CO₂ and O₃, reflecting inputs associated with combustion-derived CO₂ and pollution-derived O₃, but the corrections are relatively small (e.g., ~0.6 ppm for CO₂).

Table 4. $\langle Dq \rangle^\circ \langle (q_b - q_h) \rangle$ for different data treatments

DO ₃			
Altitudes (m)	All data	Linear Mdl _[COobs]	Linear Mdl _[CObkgd]
0 – 3300	-5.40	-7.73	-8.24
0 – 2500	-4.48	-4.80	-5.05

CO₂ and DCO₂

Treatment	q _{h=3300}	q _{b (0-3300)}	Dq ₃₃₀₀	q _{h=2500}	q _{b (0-2500)}	Dq ₂₅₀₀
"All data"	347.80	347.79	-0.01	347.45	347.85	0.40
Linear Mdl _[COobs]	346.97	347.80	0.83	347.51	347.84	0.33
Linear Mdl _[CObkgd]	346.85	347.10	0.26	347.36	347.02	-0.33

Units: CO₂, ppmv; O₃, ppbv. "All data": block averaged data by hour and 200 m altitude intervals (no linear model); "Linear Mdl", Eq. (2) with CO = block-averaged (time/height) mean; "Linear Mdl, CO_{bkgd}", Eq. (2) with CO set to background (84 ppb).

Despite the large diurnal variation in the lower part of the profile, results for $\langle(CO_2)_b - (CO_2)_h\rangle$ were surprisingly close to zero for both values of h , 0.6 ± 0.4 ppm without compensating for CO, and -0.03 ± 0.4 ppm when the covariance with CO was removed. The estimated uncertainty for this result accounts for ± 0.3 ppm from the central values in Table 4, plus the uncertainties for the average column amount (see discussion of Figure 5). An additional bias error of ± 0.3 ppm might result from diurnal bias, if our assignments of turning points were off by $\pm 1/2$ hour.

The null result for $\langle(CO_2)_b - (CO_2)_h\rangle = 0 \pm 0.7$ ppm (with conservative uncertainty, i.e. adding random and possible bias errors) contrasts markedly with results for O₃ (-6.4 ± 2.5 ppb, Table 4). The result also contrasts with CO₂ data over the US in summer [Gerbig *et al.*, 2001], where the atmosphere clearly reflected the activity of surface vegetation. Over well-watered forests, lower atmosphere was depleted by -5 to -16 ppm CO₂, whereas in drought-impacted areas CO₂ was in excess by 4-10 ppm. The US data support our concept that net sources or sinks of CO₂ should emerge as detectable contrasts between daily-average mean concentrations below h and above h . We conclude from the ~ 0 contrast over Amazônia that the CO₂ budget was very close to balance in April 1987.

Without tower data to define CO₂ at the canopy height, we would have computed a value of q_b lower by 0.1 – 0.2 ppm. This is the effect of the "diurnal

rectifier" [Denning *et al.*, 1995]. The "rectifier" was small over Amazônia in April, 1987, comparable to the uncertainty in PBL hourly values. Nevertheless it represents a potential source of systematic bias and should be accounted.

The hourly change in CO₂ column amount provides a direct measurement of regional daytime CO₂ uptake, -6.3 $\mu\text{mole m}^{-2}\text{s}^{-1}$ for both h=2500m and h=3300m, slightly lower than daytime fluxes at Manaus (average -10.2 $\mu\text{mole m}^{-2}\text{s}^{-1}$ [Fan *et al.*, 1990]). Fluxes at the tower showed ~30% greater uptake in the morning than the afternoon, due to cloudiness and stomatal closure in the afternoon, but aircraft data were more symmetrical. A likely explanation for the difference is that the aircraft observed CO₂ from wetlands, rivers, and inundated forest, which emit CO₂ and may significantly affect the regional CO₂ balance. The tower fetch did not include significant areas of inundation or wetlands.

The net 24-hour biotic exchange fluxes for CO₂ appear to be quite small, -0.13 or +0.07 $\mu\text{mole m}^{-2}\text{s}^{-1}$ for h=2500 or 3300 m respectively, using Eq. (1c) and results for O₃ (taking ΔO_3 from Table 4, O₃ flux from Fan *et al.* [1990]). Daily mean uptake corresponding to an annual rate of only -0.25 $\mu\text{mole m}^{-2}\text{s}^{-1}$ (1 ton C ha⁻¹yr⁻¹) would correspond to ΔCO_2 of -0.8 ppm, at the extreme end of our error bounds for ΔCO_2 with additional allowance for uncertainty due to lack of perfect similarity between CO₂ and O₃ and uncertainty in the mean O₃ flux over Amazônia. The net biogenic CO₂ flux was apparently less than this rather small flux.

Concentrations of CO were much lower than in ABLE-2A. If we assume that most CO enhancements over background were combustion-related, we can compute the associated CO₂ flux from the region (comparing ΔCO_2 for CO_{obs} versus CO_{bkgd}). Biomass burning fluxes for CO₂ were modest (+0.25 $\mu\text{mole m}^{-2}\text{s}^{-1}$), consistent with low rates of biomass burning in the wet season, but nevertheless exceeded regional biogenic uptake.

We estimated the fetch for the aircraft measurements using ozone data combined with rawinsonde measurements, as summarized in Figure 6. Six stations made four soundings daily in and around the Basin. The density-weighted mean wind for 0 – 3.3 km averaged 6.4 m s⁻¹ from the ESE in the main area of aircraft operations (station EMBRAPA), with similar values upwind at Belém and Alta Floresta. The coast is about ~2400 km ESE of our operational site, giving an average transit time, t_{tr} , of 4.3 days. If we assume that the O₃ profiles satisfied the quasi-periodic condition, we infer $\tau_{exch} = 2.7$ days from Eq. (1b) and Table 4. Pereira [quoted by Jacob and Wofsy, 1990] measured ²²²Rn on the Electra, and inferred $\tau_{exch} \sim 3$ days for the PBL, in harmony with the value derived here. The observation that the transit time was longer than τ_{exch} lends support to our adoption of the quasi-periodic approximation in our simple analysis of concentration gradients and fluxes.

The values of t_{tr} and τ_{exch} imply a fetch of roughly 1500 km, extending over the equatorial forest from west of Manaus to southwest of Belém. This is only a rough indication of the area sampled, since wind speed increases with altitude. It seems clear however that the aircraft data characterized a vast region of mostly intact equatorial forest and associated mosaics of wetlands, inundated lands (*varzea*, *igapo*), and great rivers (20-30% of the land area), at a time of high, and increasing, water levels.

Discussion

Grace et al. [1996] derived annual mean NEE of $-8.5 \text{ mole C m}^{-2}\text{yr}^{-1}$ ($-0.3 \text{ } \mu\text{mole m}^{-2}\text{s}^{-1}$) from eddy covariance measurements in Rondônia (10°S , 57°W). *Malhi and Grace*, [2000] argued that intact tropical forests globally represent a sink for CO_2 of 2.0 PgC yr^{-1} , stimulated by rising atmospheric CO_2 that could offset 80-90% of the CO_2 source from deforestation. Most of this sink should lie in Amazônia, which has ~60% of lowland tropical forest globally, and a larger fraction of intact tropical forests. If this mean rate were effective in April 1987 in Amazônia, we should have seen a negative CO_2 gradient close to 1 ppm that was not observed.

Model results [Figure 7, *Botta and Foley*, 2001] indicate that forest growth should have been near a seasonal high. The model computed net exchange of CO_2 for Amazônia from 1935 to 1995, using recorded weather data and the Integrated Biosphere Simulator (IBIS) [*Foley et al.*, 1996; *Kucharik et al.*, 2000] to compute the energy, water, and carbon balances of the land surface, plant physiological processes (photosynthesis, respiration), phenology, plant growth and competition, vegetation dynamics, and nutrient cycling. The uncertainty bars in Figure 7 show the standard deviation of model fluxes for nine $1^{\circ}\times 1^{\circ}$ grid squares in the fetch. *Botta and Foley* [2001] found uptake in April-May 1987 slightly greater than the median for April-May in all years, reflecting somewhat drier conditions than average (more sunshine). Release of CO_2 due to drought was predicted later in 1987 [Cf. *Tian et al.*, 1998], well after ABLE-2B.

Our observed value is at best marginally consistent with model results in Fig. 7. Since deforestation and biomass burning are both at seasonal lows late in the wet season, and drought had not yet begun, there appear to be neither climatic reasons nor disturbance scenarios to explain the zero flux observed in April 1987.

Rivers and flooded lands are large, persistent sources of CO_2 to the atmosphere [*Wofsy et al.*, 1988; *Richey et al.*, 1990], and a significant quantity of this CO_2 arises from organic matter transported to watercourses from *terra firme* forests and marginal wetlands [*Quay et al.*, 1992]. Tower studies and ecological models generally focus on forests in upland areas, whereas our aircraft data integrate over the landscape. Thus we should have expected regional fluxes to be more positive than fluxes from towers or models. In fact, *regional* decay rates likely peak late in the wet season, due to hydrological factors not usually modeled: expanding areas of inundation, overland flow of organic matter to water courses, and increased soil moisture lead to higher rates of decomposition. A plausible explanation for lack of CO_2 uptake in ABLE-2B is that forest growth was masked by seasonal enhancement of decomposition. Hence it is important to account for seasonal variations of both growth *and* decomposition and to examine linkages between components of the landscape mosaic.

Conclusions

ABLE-2 provided the first comprehensive look at temporal and spatial distributions of CO_2 and other tracers in the tropical atmosphere. We analyzed the historical data for CO_2 in ABLE-2B to show that regional fluxes for CO_2 lead to atmospheric concentration gradients below 3 km that can be quantified by systematic

aircraft soundings. We provided a first-order assessment of regional net fluxes for April 1987 by complementing aircraft data with tracer and meteorological data, and with measurements from flux towers.

Significant covariance was observed between CO₂ and CO, due to the influence of surface sources and large-scale mixing, requiring careful analysis to extract information on regional sources and sinks. The relationship between diurnal variations of CO₂ and convective rainfall was more symmetrical than anticipated, giving rise to only small gradients due to temporal covariance of fluxes and convective overturning ("rectification"). Concentrations of O₃ were strongly depleted in the lower atmosphere due to uptake by the vegetation, a well-characterized process in terms of deposition velocity and flux. Since photochemical rates are very slow in the wet season, net fluxes of CO₂ could be estimated by similarity with O₃.

The layer-mean (0 to $h=3300\text{m}$ or 2500m) concentration of CO₂ declined by 3-4 ppm from morning to evening, but the average value was almost exactly the value observed just above h . We infer that rates for daytime uptake were large, but there was no observable *net* uptake or release of CO₂ over 24 hours. The fetch for these measurements, ~1500 km, encompassed a large fraction of the equatorial Amazon Basin. The lack of regional uptake for CO₂ in the latter stages of the wet season was surprising given the strong ecological reasons to expect net growth of forest trees at this time, and it was in sharp contrast with aircraft flights over the northern US in August, 2000. In the US, uptake of CO₂ by forests drew down 24-hour mean concentrations in the lower atmosphere by 3-4 ppm.

Our analysis draws attention to a critical difference between studies at ecosystem scales, such as flux towers and ecosystem models, versus regional scales. In Amazônia, up to 30% of the land surface may be inundated late in the wet season, providing strong sources of atmospheric CO₂. Much of the carbon released from these areas is derived from forests. Hence the regional CO₂ flux from Amazônia to the global atmosphere may be quite different than the exchange associated with an individual component, even a major component such as *terra firme* forests. Measurements of large scale landscape mosaics are required in order to determine the carbon budget for a region or continent, and should provide important input for global inverse-model studies. ABLE-2B data provided a preliminary set of large-scale measurements to examine and interpret for these purposes. More rigorous continental fluxes can be obtained if observations are made along with detailed meteorological observations, and analyzed using the advanced mesoscale data assimilation models becoming available at the present time.

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