

## Net fluxes of CO<sub>2</sub> in Amazonia derived from aircraft observations

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[1] A conceptual framework is developed using atmospheric measurements from aircraft to determine fluxes of CO<sub>2</sub> from a continental land area. The concepts are applied to measurements of CO<sub>2</sub>, O<sub>3</sub>, and CO concentrations from the Amazon Boundary Layer Experiment (ABLE-2B, April–May 1987) to estimate fluxes of CO<sub>2</sub> for central and eastern Amazonia late in the wet season of 1987. We observed that column amounts of CO<sub>2</sub> from 0 to 3 km decreased during the day over Amazonia at the average rate of  $-6.3 \pm 1 \mu\text{mol m}^{-2} \text{s}^{-1}$ , corresponding to an uptake flux modestly smaller than the daytime uptake ( $-10.2 \mu\text{mol m}^{-2} \text{s}^{-1}$ ) at a flux tower in the study area. The estimated net flux of CO<sub>2</sub>, integrated over 24 hours, was  $-0.03 \pm 0.2 \mu\text{mol m}^{-2} \text{s}^{-1}$ , indicating that the carbon budget of a substantial area of central Amazonia was close to balance in April 1987. We argue that net CO<sub>2</sub> fluxes on the continental scale of Amazonia, with its heterogeneous landscape and large areas of inundation, are strongly modified by the influence of seasonal hydrological factors that enhance respiration and decomposition in forests and wetlands, offsetting growth of forest trees in the wet season. *INDEX TERMS:* 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 1610 Global Change: Atmosphere (0315, 0325); *KEYWORDS:* carbon cycle, Amazon Basin, aircraft measurements

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### 1. Introduction

[2] Global studies of atmospheric CO<sub>2</sub>, O<sub>2</sub>, and <sup>13</sup>CO<sub>2</sub> 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] proposed that mature Amazonian forests may be major sinks for CO<sub>2</sub>, up to 2 Pg C/yr, based on data from ecological plots and eddy correlation flux towers. They cited increasing CO<sub>2</sub> concentrations as a possible stimulus for carbon uptake by tropical forests.

[3] Conflicting claims for carbon sinks are difficult to test due to lack of data over the continents. Most atmospheric CO<sub>2</sub> observations 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. Since few data exist over the continents, a priori specifications of flux patterns over the surface play an important role in inverse models [Kaminski and Heimann, 2001; Rayner *et al.*, 1999]. Large-scale atmospheric data are needed to evaluate these models and to test inferences from plot-size data indicating uptake of CO<sub>2</sub> by mature tropical forests.

[4] The present paper introduces a new conceptual framework for using concentration data from aircraft to estimate regional fluxes of CO<sub>2</sub> over a continent. We use mass balance calculations for the atmospheric column for each hour of the day, and account for CO<sub>2</sub> from combustion and from long-range transport using CO data. The concepts are applied to historical data acquired in Amazonia during the Amazon Boundary Layer Experiment (ABLE-2B) campaign. Our approach derives values for the rate of daytime uptake by the mosaic of surface vegetation and estimates the regional net 24-hour flux of CO<sub>2</sub>. The work extends analysis of column integrals presented for ABLE-2A from 1985 [Wofsy *et al.*, 1988]. The results suggest that the net flux for CO<sub>2</sub> was  $\sim 0$ , averaged over a substantial part of Amazonia, in April 1987 [Chou, 1999].

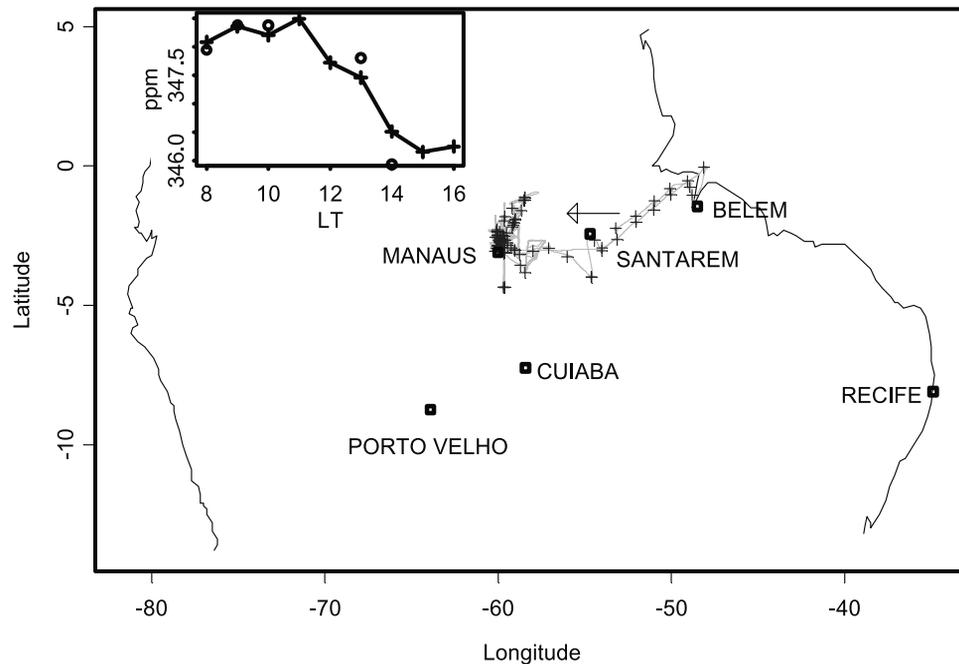
### 2. Observations

[5] ABLE-2B was conducted in April and May 1987, months 6 and 7 of the 9-month wet season. Instruments on board the NASA Electra aircraft measured CO<sub>2</sub>, O<sub>3</sub>, CO, NO, aerosols, and meteorological parameters for 21 mis-

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**Figure 1.** Map showing locations (pluses) for the 85 vertical profiles acquired during ABL-2B, 15 April–8 May 1987. The wind in the lower 3 km of the atmosphere averaged  $6.4 \text{ m s}^{-1}$  from the East (arrow), giving a transit time of 4.3 days to the main sampling area northeast of Manaus, 2400 km from the ocean. (inset) Diurnal variation of the mean column concentration of CO<sub>2</sub> (0.2–2.8 km) in central Amazonia (pluses, west of  $-56^\circ$ ) compared to values in eastern Amazonia (circles, east of  $-56^\circ$ ). See color version of this figure at back of this issue.

sions of four types [Harriss *et al.*, 1990a]: Surveys flew east from Manaus to Belém and back (Figure 1). Source missions intensively sampled the lower atmosphere (0–3 km) over selected vegetation, such as forest or wetland, principally north of Manaus. Convective transport flights covered a wide altitude range to examine the influence of convection. A 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).

[6] Data for CO<sub>2</sub> were obtained on 15 missions spanning hours from 0700 to 1700 local time (denoted LT, i.e., GMT - 4 hours) over 24 days, at altitudes from 0.15 to  $\sim 6$  km. Included were: seven source missions (one principally over wetlands) that repeatedly sounded the atmosphere in one area; four transport missions attempting to bound a “volume” and to measure advection and divergence of tracer; three survey missions taking vertical profiles across the Basin; and the flux mission following a racetrack pattern in and just above the Planetary Boundary Layer (see Table 1 and Chou [1999]). Most data were acquired between 0.15 and 3.1 km.

**Table 1.** Electra Aircraft Missions During ABL 2B

Flight	Local Time	Day of 1987	Mean Latitude, °S	Mean Longitude, °W	Mission Type <sup>a</sup>	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-Belém survey	0.2–3.0
12	7–11	114	NA	51	Belém-Santarém survey	0.2–3.0
13	14–17	114	3.0	58	Santarém-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

<sup>a</sup> Browell *et al.* [1990].

[7] Concentrations of CO<sub>2</sub> 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 CO<sub>2</sub> concentration 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 ~200 m in the horizontal and ~10 m in the vertical.

[8] Calibrations were carried out frequently in flight by flowing standard gases (CO<sub>2</sub> 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.

[9] Corrections were applied to account for BINOS sensitivity to aircraft attitude and acceleration by fitting data from calibrations to a second-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.

[10] Concentrations of O<sub>3</sub> were measured using ethylene 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]. Concentration data, including CO, O<sub>3</sub>, and CO<sub>2</sub>, were averaged into 10 s intervals and archived. We used archived data to create a merged data set. The instruments did not report concentrations during eddy flux measurements, since accurate calibrations were impossible due to high flow rates.

[11] Additional measurements for CO<sub>2</sub> and O<sub>3</sub> were obtained at a tower in Ducke reserve on the outskirts of Manaus, including eddy covariance fluxes at 41 m and concentrations at 0.02, 3, 6, 12, 27, 36, 41 m [Fan *et al.*, 1990]. Results were reported for ~8 days with mostly non-precipitating conditions. The fetch for this tower was predominantly upland forest without large watercourses or extensive wetlands.

[12] Figures 2a and 2b show data from an illustrative subset of flights. Flight 14 sampled over forests in Central Amazonia northwest of Manaus, flight 12 was a transit flight in eastern Amazonia between Santarém and Belém, and flight 18 measured a mix of wetlands and forests near Manaus. Combustion plumes were observed on flight 14

(note spikes in CO at 7.9, 9.4, and 9.7 hours in Figure 2a), possibly emanating from Manaus. Weaker emissions from biomass fires were seen on many flights. CO<sub>2</sub> data from eastern and central Amazonia were very similar (Figure 1, inset). 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, upper panels). Elevated CO<sub>2</sub> is seen at low altitudes in the morning, as CO<sub>2</sub> respired at night mixes upwards into the growing PBL. Low CO<sub>2</sub> just above the morning PBL (0.6–1.2 km) arises from the relict PBL of the previous afternoon, where low CO<sub>2</sub> concentrations (due to photosynthesis) are preserved absent vertical mixing during the night. Small drawdowns were observed consistently in the afternoon, as seen, for example, on flight 18 (Figure 2b, lower panel). Observed differences between CO<sub>2</sub> in the relict PBL and air aloft ( $\geq 3$  km) were much smaller in Amazonia (3–5 ppm) than in North America in summer (10–20 ppm or more) [Gerbig *et al.*, 2001].

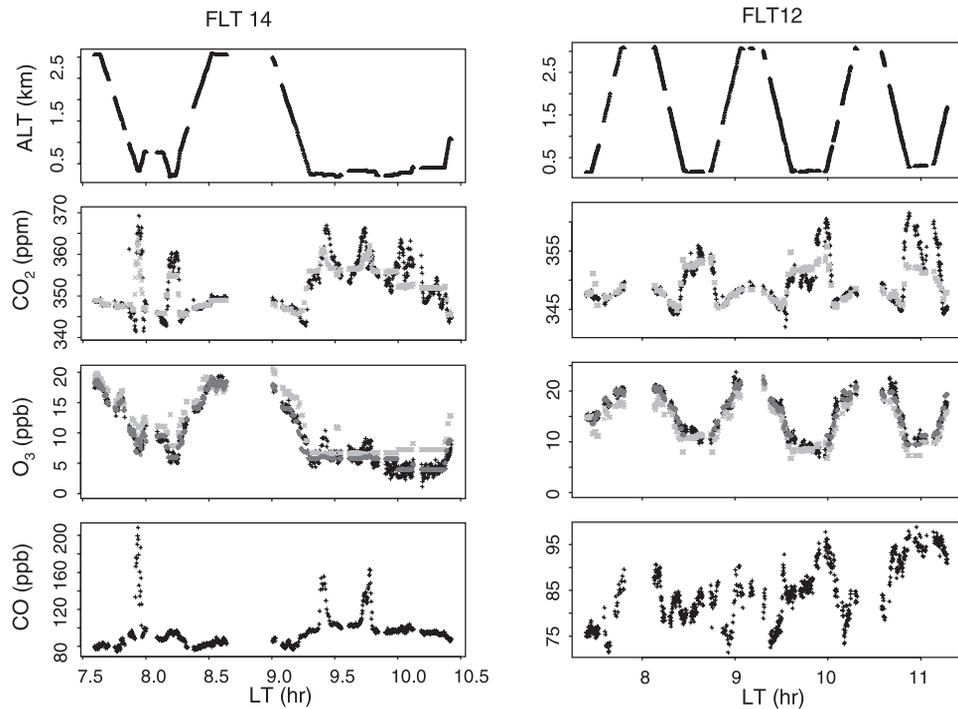
[13] Figures 3a–3c show mean vertical profiles of CO<sub>2</sub>, CO and O<sub>3</sub> block averaged by hour and altitude (200 m bins) over the 24 days/15 flights; tower data block-averaged by height and hour of the day were appended at the bottom. The lower atmosphere steadily loses CO<sub>2</sub> during the day due to photosynthetic uptake, although midday CO<sub>2</sub> concentrations remain higher than aloft. Late in the afternoon PBL values were depleted by 1–2 ppm compared to air higher up (Figure 3a). Data for 1400 Local Time are anomalous above ~1.5 km, reflecting unusual CO, O<sub>3</sub> and CO<sub>2</sub> data from a single flight (flight 7), which we judge to be unrepresentative. Apart from these data on flight 7, the flights were remarkably consistent despite fine-scale variance.

[14] The CO<sub>2</sub> profile averaged over the day shows a weak minimum between 1 and 2 km, altitudes where exchange with the surface takes place mainly in the afternoon when concentrations are low. There is a corresponding enhancement of daily mean CO<sub>2</sub> at 500 m and below (Figure 3a). This contrast (“rectification”) arises from the correlated response of photosynthesis and of growth of the PBL to diurnal forcing [Denning *et al.*, 1995, 1999]. The magnitude (~1 ppm) and vertical extent (~2 km) were smaller in ABLE-2B than 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, notably different from results at midlatitudes [Gerbig *et al.*, 2001], as discussed in detail below.

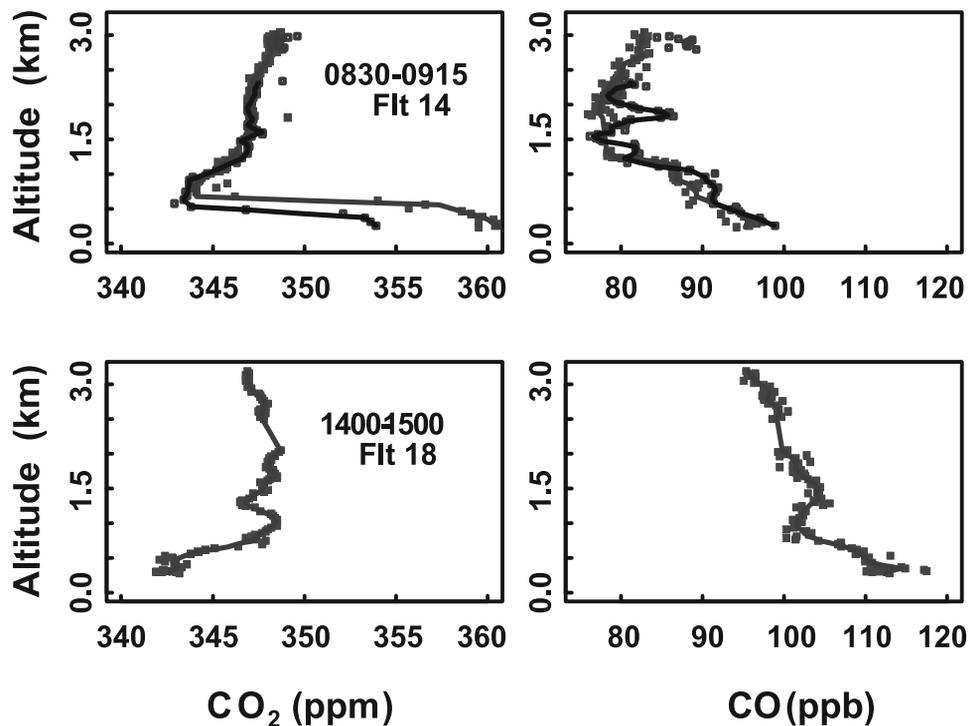
[15] Concentrations of CO were generally highest near the ground, and typically increased during the day at low altitudes (Figures 3b and 3d), due to inputs from the surface. Concentrations of O<sub>3</sub> were ~20 ppb at 2 km, declined to 10 ppb at the canopy, and vanished at the ground, due to strong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

### 3. Framework for Analysis of Aircraft Measurements

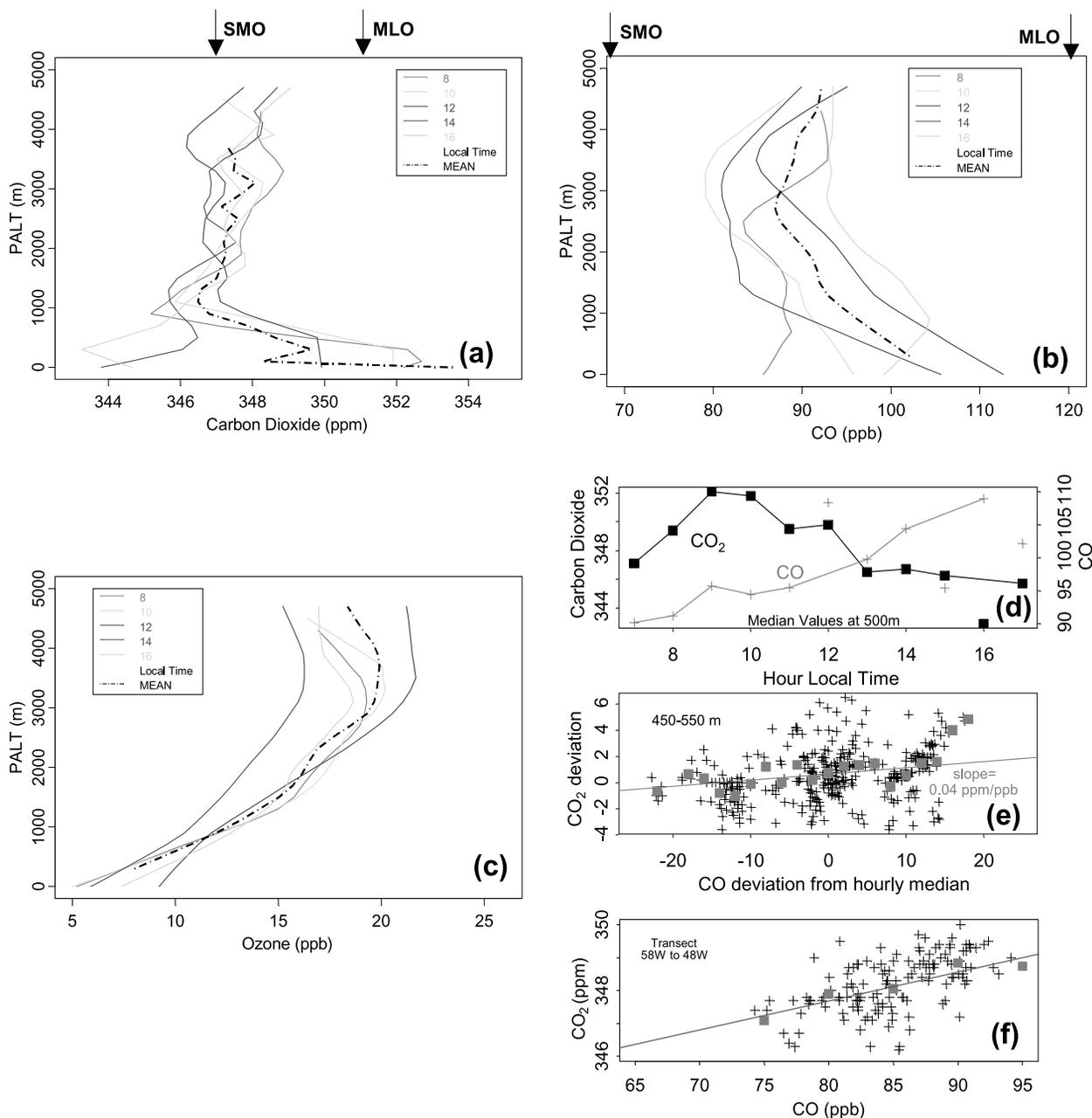
[16] We wish to derive mass budgets for atmospheric CO<sub>2</sub> and O<sub>3</sub> for the continental region upwind of our observa-



**Figure 2a.** Data for altitude, CO<sub>2</sub>, O<sub>3</sub>, and CO are plotted versus time for flights 12 and 14 (see Table 1). Black points show the observations for 10 s intervals with valid data for CO<sub>2</sub>. Blue points show linear model representation of the data using CO as a predictor with hour and altitude as factors. Red points for O<sub>3</sub> add date as a factor to account for variation of background O<sub>3</sub> concentrations. Smoke layers encountered on flight 14 (8, 9.5, 9.7 hours) contributed up to 15 ppm CO<sub>2</sub>. See color version of this figure at back of this issue.



**Figure 2b.** Vertical profile data (points) for CO<sub>2</sub> and CO for flight 14 (upper panels) and flight 18 (lower panels), and smoothed curves from locally weighted least squares (lines). Note the close agreement of the two profiles (30 minutes apart) above the PBL for both CO<sub>2</sub> and CO in flight 14. Data gaps show locations of midprofile calibrations.



**Figure 3.** (a) Vertical profiles for CO<sub>2</sub> from 15 flights, days 105–126, 1987, in central Amazonia (Table 1), block averaged by local time. (b) Vertical profiles for CO, as in Figure 3a. Arrows show data from mid-Pacific stations Samoa (SMO, 14° 15'S, 170° 34'W) and Mauna Loa (MLO, 19° 32'N 155° 35'W) for April 1987 (CO<sub>2</sub>) and for April 1990 (the first year of station data for CO). (c) Vertical profiles for O<sub>3</sub>, as in Figure 3a. (d) Diurnal variation of CO<sub>2</sub> and CO concentrations at 500 m (median values for each hour, all flights). Lines illustrate the general trends during the day. (e) Deviations of CO<sub>2</sub> and CO from the hourly median concentrations (Figure 3d) at 500 ± 50 m. (f) CO and CO<sub>2</sub> gradients across the Amazon Basin (Santarém-Manaus survey) at 3 km altitude: pluses, raw; minuses, block averaged by CO (1 ppb bins, Figure 3e; 5 ppb bins, Figure 3f), linear regression line; straight-line connecting SMO and MLO data points (slope = 0.08 ppm/ppb). See color version of this figure at back of this issue.

tions, from the ground to fixed height  $h$ . Ideally, we would use a high-resolution mesoscale model to derive rates of mass transport, including detailed representation of convection, but unfortunately this is impossible for our historical data set. Hence we introduce a simple conceptual model representing the basic elements of transport, to elucidate the

observational strategies needed to determine continental-scale fluxes from aircraft data. We also examine the influence of fluxes with a diurnally varying component on the near-surface anomalies in tracer concentrations.

[17] Air enters the Amazon Basin with the Trade Winds from the east (Atlantic Ocean) and flows towards a major

region of convergence in western Amazonia. The observed winds were almost due easterly from  $\sim 1$  to 10 km throughout the equatorial zone, with maximum speed of  $10 \text{ m s}^{-1}$  at 2 km (see Figure 7a below), turning more northerly right near the surface. Flights in ABLE-2B stayed upwind of the main convergence zone, but mostly downwind of the equatorial forests that lie between Manaus and the ocean [Santos, 1987]. Rainfall averaged 200.3 mm during the study [Garstang *et al.*, 1990], ample, but below the long-term mean ( $\sim 330$  mm). Despite significant rainfall, Central and Eastern Amazonia experienced net divergent flow [Greco *et al.*, 1990], providing weak net subsidence ( $\sim 200$  m/day at 700 mb) over the Basin. Convection was limited to small, local storms on 42% of the study days, with moderate-scale, early-morning systems (“basin occurring systems”) on 30% of the days. Typical PBL developments, responding to solar heating during the day, were observed over the Basin on these days, accounting for more than 70% of study interval [Greco *et al.*, 1990].

[18] Our simple concept envisions 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 throughout the PBL and associated Convective Cloud Layer (CCL), while exchanging slowly 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, namely, up to the maximum daily height,  $h$ , of the PBL plus CCL. Thus the chemical composition of air below  $h$  is controlled by surface fluxes and by fluxes across  $h$ , averaged over a diurnal cycle. In contrast, air above  $h$  reflects the large-scale circulation. We select a fixed value for  $h$  to include all altitudes in daily contact with the surface, on days with well-defined growth and decay of the PBL, using meteorological and chemical measurements. We considered bounding values (2500 m and 3300 m) to insure that results were not significantly affected by the choice of  $h$ .

[19] The concepts are expressed mathematically by vertically averaging the mass continuity equation for tracer in a column with unit area and height  $h$ , transported with the mean flow,

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

Here  $S_i$  denotes surface flux for one of the species for this study, CO<sub>2</sub> or O<sub>3</sub> ( $\text{mol m}^{-2} \text{ s}^{-1}$ ),  $[P_i - L_i]$  is the net chemical tendency averaged over altitudes  $0 - h$  ( $\text{mol m}^{-3} \text{ s}^{-1}$ ),  $q_{ib}$  is the mean mole fraction of tracer  $i$  between 0 and  $h$  ( $q_{ib} \equiv \frac{1}{n_b} \int_0^h q_i n dz$ ),  $n_b$  ( $\equiv \frac{1}{h} \int_0^h n dz$ ) is the mean atmospheric number density ( $\text{mol m}^{-3}$ ) 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  $hn_h/\tau_{\text{exch}}$  is the mean mass flux of air across  $h$ . Time  $t$  is measured from the entry of air onto the continent. The “column-averaged net source” ( $S_i/h + [P_i - L_i]$ ) combines the effects of surface flux and chemical reactions on the mean atmospheric composition for altitudes 0 to  $h$ . The chemical tendency  $[P_i - L_i]$  is zero for CO<sub>2</sub>, and negligibly small for O<sub>3</sub> under the conditions for ABLE-2B

(see below). The level  $z = 0$  is set at top of the forest canopy, since aircraft data do not resolve CO<sub>2</sub> sources or concentration gradients within the forest.

[20] The main assumption made in equation (1a) is that vertical exchange via cloud venting and subsidence can be represented by a time independent value for  $\tau_{\text{exch}}$ . If vertical exchange through  $h$  is not strongly biased towards particular hours of the day, the simple parameterization in equation (1a) does not lead to a bias that can affect inferred values of the net surface source (see Appendix A). The observed pattern of deep convective rainfall in ABLE-2B confirms that  $q_{ih}$  may be assumed time-independent. Hence equation (1a) may be rewritten as a differential equation for the difference between  $q_h$  and the concentration averaged over the column  $0-h$ ,  $\Delta q_i$  ( $\equiv q_{ib} - q_{ih}$ ), in terms of  $S_i$  and the mean replacement time for atmospheric mass between 0 and  $h$ ,  $\tau \equiv \tau_{\text{exch}} n_b/n_h$ ,

$$\frac{\partial \Delta q_i}{\partial t} + \frac{\Delta q_i}{\tau} = \frac{S_i}{n_b \cdot h}. \quad (1b)$$

[21] We may divide  $S_i$  into a periodic part,  $S'_i$  (period  $T = 1$  day, zero mean over 24 hours) plus the 24-hour mean net flux,  $\bar{S}_i$  both assumed invariant over the footprint. The solutions  $\Delta q_i$  must likewise have a periodic part,  $\Delta q'_i$  with zero mean over 24 hours, and a non-periodic part,  $\overline{\Delta q_i}$ . Invariance of  $S_i$  over the fetch for the flights appears to be an excellent approximation in light of the similarity of average concentrations and diurnal variations in the layer  $0-h$ , from Manaus eastward almost to Belém (Figure 1, inset), as observed in our cross-Basin flights.

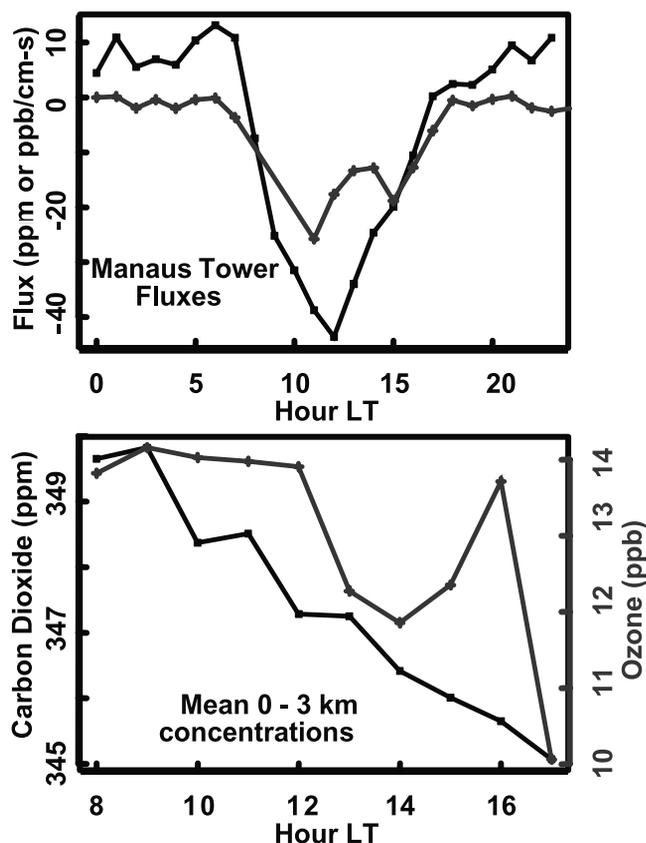
[22] Appendix A provides the solutions to equation (1b) for these assumptions. The 24-hour average of  $\Delta q_i$  (defined as

$$\frac{1}{T} \int_t^{t+T} \Delta q_i dt,$$

and denoted by the overbar) is a direct measure of the 24-hour net surface flux (equation (A2)),

$$\overline{\Delta q_i} = \frac{\bar{S}_i \tau}{n_b h} \left( 1 - e^{-t/\tau} \right), \quad (1c)$$

where tracer concentrations are assumed well mixed over the ocean ( $q_{ib} = q_{ih}$ ) <sub>$t=0$</sub> , as observed in ABLE-2A [Wofsy *et al.*, 1988]. Equation (1c) shows that the 24-hour mean column concentration uniformly approaches a steady state that depends only on the 24-hour mean surface flux. The rate of approach depends on the timescale for entrainment from aloft. The 24-hour mean surface flux can thus be determined from the 24-hour mean difference in concentrations between the free troposphere and the lowest layer of the atmosphere (between 0 and  $h$ ). Periodic (diurnal) changes in concentrations, and associated diurnally varying fluxes, are separable and may be determined independently. These relationships arise from elementary considerations of mass balance, and are more general than the simple derivation given in Appendix A.



**Figure 4.** (top) Fluxes of CO<sub>2</sub> (black) and O<sub>3</sub> (gray) versus local time at the Manaus tower [Fan *et al.*, 1990]. (bottom) Density-weighted mean concentrations during the day for the column 0–3.1 km from the aircraft. See color version of this figure at back of this issue.

[23] Equation (1c) implies that there is a similarity relationship for the 24-hour mean concentrations and surface fluxes of two tracers,

$$\frac{\overline{\Delta q_1}}{\overline{\Delta q_2}} = \frac{\overline{S_1}}{\overline{S_2}} \quad (1d)$$

That is, 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  $h$  ( $q_b$ ),  $\Delta q_i$ , also averaged over 24 hours. It is not necessary to know dynamical quantities such as  $\tau$ . Equation (1d) holds if we sample air parcels with an ensemble of ages, for example due to horizontal dispersion within the Basin, even though equation (1c) would be problematic. The main requirements are separation of diurnal from longer timescales, for both the surface forcing and replacement of PBL air, and linearity of the exchange processes. Equation (1d) is the key relationship we will use to estimate regional fluxes.

[24] In ABLE-2B we obtained data for daytime hours from 7 to 17 hours local time, with a few after 17 hours. The observations confirm that concentrations in the lower atmosphere are quasi-periodic, implying that increases in CO<sub>2</sub> at night must, on average, reverse the daytime draw-

down. Our data cover the daily turning points of the column concentrations of CO<sub>2</sub>, observed at ~8 hours (daily maxima) and between 16 and 17 hours (minima) in ABLE-2A [Wofsy *et al.*, 1988], when nighttime data were obtained. The turning points are consistent with times for reversal of the CO<sub>2</sub> flux at the tower (Figure 4 [Fan *et al.*, 1990]). Flux reversals occurred when photosynthesis just balanced soil respiration, about 1 hour after sunrise and 1–2 hours before sunset. Thus our daytime data in ABLE-2B are sufficient to derive the 24-hour mean values for  $q_{ib}$  required for equation (1d).

[25] The average daytime value of  $S_i$  can separately be determined from our hourly data for  $q_{ib}$ , including the diurnally varying components of  $S_i$ ,  $\langle S_i \rangle_{\text{day}} = n_b h \langle \partial \Delta q_i / \partial t \rangle_{\text{day}}$  (see Appendix A), where

$$\langle \dots \rangle_{\text{day}} \equiv \frac{2}{T} \int_{T/4}^{3T/4} \dots dt$$

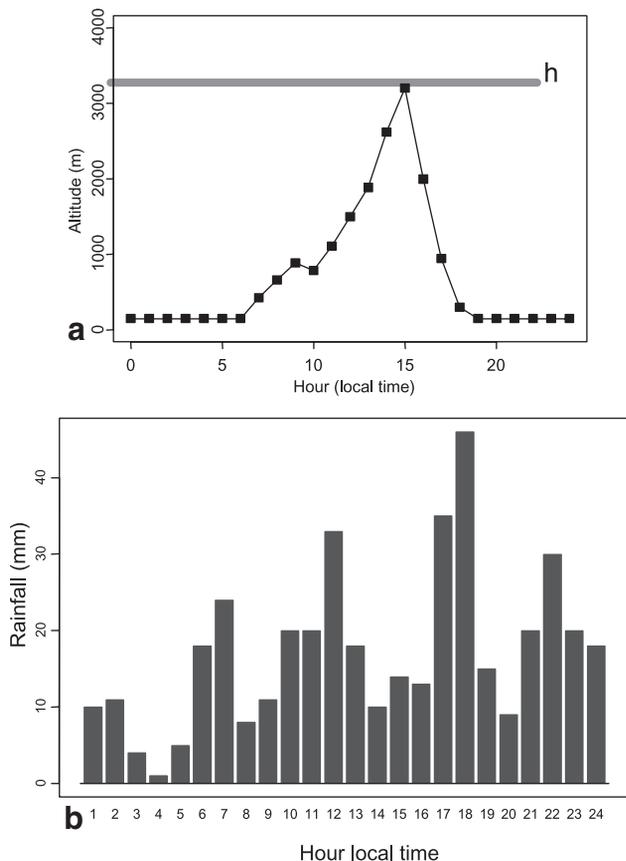
is the average value over the daytime hours. These results may be compared to tower data for daytime uptake of CO<sub>2</sub> by the forest [cf. Wofsy *et al.*, 1988].

[26] Our analysis of column budgets using equations (1a)–(1d) 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  $q_{ib}$  in (1a) 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.

[27] The height of the fixed level  $h$  (Figure 5a) was selected by examining profiles for CO, O<sub>3</sub>, and H<sub>2</sub>O for each flight to estimate the altitudes influenced by surface exchange, i.e., the height of the PBL plus 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 CO<sub>2</sub> 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.

[28] Since CO<sub>2</sub> is inert in the atmosphere, but ozone is not, similarity requires that the chemical tendency for O<sub>3</sub> ( $P - L$ ) in the atmosphere be small compared to the surface flux  $S/h$ . Fluxes of O<sub>3</sub> to the forest were  $-3.8$  and  $-0.37$  nmol m<sup>-2</sup> s<sup>-1</sup> in the day and night, respectively (Figure 4, upper panel; Fan *et al.* [1990]), giving  $\overline{S_i}/h = -6.8 \times 10^{-13}$  mol m<sup>-3</sup> s<sup>-1</sup> for  $h = 3300$  ( $-8.4 \times 10^{-13}$  mol m<sup>-3</sup> s<sup>-1</sup> for  $h = 2500$  m). Model results [Jacob and Wofsy, 1990] indicated that the chemical tendency for O<sub>3</sub> averaged  $2 \times 10^{-14}$  mol m<sup>-3</sup> s<sup>-1</sup>, less than 3% of the deposition flux, due to very low concentrations of NO. Hence chemical reactivity was insignificant for O<sub>3</sub>.

[29] Fine-scale variations of CO<sub>2</sub> were consistently correlated with concentrations of CO (Figures 2a and 2b), especially in distinct concentration spikes (e.g., Figure 2a, flight 14). Some of this covariance was associated with combustion sources within the study area, but some arose from large-scale (e.g., interhemispheric; Figure 3f) transport. Concentrations of both CO<sub>2</sub> 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



**Figure 5.** (a) Mean diurnal variation of the height influenced by exchange with the surface inferred from aircraft profiles of CO<sub>2</sub>, O<sub>3</sub>, H<sub>2</sub>O, and temperature. The height  $h$  in equation 1 is indicated by the grey horizontal line. (b) Mean hourly rainfall from a basin-wide network of automated weather stations [from *Greco et al.*, 1990].

in CO<sub>2</sub> and CO from W to E across the Amazon Basin closely approximated a mixing line with end-members at SMO and MLO (Figures 3a, 3b, and 3f). These data are consistent with the observation [*Boering et al.*, 1994; *Andrews et al.*, 1999] that concentrations of CO<sub>2</sub> 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  $\sim 16$  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.)

[30] Our concept requires that we distinguish changes in CO<sub>2</sub> due to forest metabolism from changes due to combustion, or from variable admixtures of Northern and Southern hemisphere air transported into the study area. We observed that combustion and large-scale mixing produced similar correlations, 0.04–0.1 ppm CO<sub>2</sub>/ppb CO [cf. *Andreae et al.*, 1988]. We therefore developed a statistical relationship between CO and CO<sub>2</sub> at each altitude to remove the influence of these processes on  $q_{ib}$  and  $q_{ih}$ . We compared the result to straightforward conditional sampling that removed high CO values, as described below.

[31] A key assumption in the derivation of equation (1d) 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  $q_{ih}$ . Another assumption is that air leaving the column has the 24-hour column mean concentration,  $q_{ib}$ . If convection occurred preferentially at times of day when concentrations of CO<sub>2</sub> in the PBL differed from the 24-hour mean value, transport through level  $h$  could be biased (see Appendix A). Fortunately the distribution of convective rainfall over the day was almost unbiased relative to the diurnal cycle of CO<sub>2</sub> (Figure 5b), and the bias in  $q_{ib} - q_{ih}$  was estimated to be at most 10% towards lower concentrations. Observed precipitation was notably less biased towards afternoon than predicted by some models [e.g., *Peylin et al.*, 1999].

#### 4. Linear Models Representing the Aircraft Data Set

[32] We sought mean profiles representing all 15 flights throughout the Basin for 24 days, to average out variations associated with weather, particular locations, instrument noise, etc., using CO as a tracer to remove the influence of combustion and of long-range advection (interhemispheric exchange). We adopted several alternative approaches to this averaging problem, to insure we did not introduce spurious results.

[33] First we defined mean profiles by block-averaging all data by hour and by altitude (200 m intervals) and then computed column-mean concentrations  $q_{ib}$  and concentrations at  $h$  ( $q_{ih}$ ) for each hour of the day. This simple procedure does not account for combustion inputs or long-range transport. Next, we used conditional sampling, removing O<sub>3</sub> and CO<sub>2</sub> data associated with CO values >90 ppb, and block-averaged the remaining data (3400 of 9400 observations) as before. Conditional sampling insures that artifacts due to combustion inputs and large-scale advection are small, but slight biases could remain. Results for these methods were very close (within 0.1–0.2 ppm) to those obtained using the more complete treatment described below, indicating that our procedures to account for systematic covariance of CO and CO<sub>2</sub> did not introduce artifacts.

[34] Our third method derived linear functions to represent data for CO<sub>2</sub> and O<sub>3</sub> 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 the “Linear Model” are

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

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

Here  $[\text{CO}_2]_j$  and  $[\text{CO}]_j$  denote observed concentrations in each 10 s interval falling in the  $j$ th altitude band at hour  $t$  (truncated to the nearest hour). The terms  $\sum_i a_{ji} \delta_{ji}$  or  $\sum_i b_{ji} \delta_{ji}$  represent mean concentrations at each altitude in the

**Table 2.** CO<sub>2</sub> Linear Model<sup>a</sup>

Altitude, m	Mean	$\sigma_r$	R <sup>2</sup>	$a_{j1}$
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

<sup>a</sup>Mean equals 24 hour average (ppm).

(*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  $b_{j0}$ ).

[35] Equation (2) with optimized coefficients provides mean CO<sub>2</sub> and O<sub>3</sub> at each altitude for each hour, and the mean dependence on CO at each altitude. We can then generate synthetic CO<sub>2</sub> profiles, in which the effects of combustion sources and advected CO<sub>2</sub> are removed by setting CO to its background concentration.

[36] If we allow for residual variance from the CO<sub>2</sub> instrument, 1–2 ppm, the Linear Model accounted for more than 60% of observed atmospheric variance up to about 1100 m, excellent fits considering the composite treatment of data spanning a month (see Table 2, and fitted points for typical flights in Figure 2a). Above ~1300 m instrument variance exceeds atmospheric variance and the fits are difficult to assess. The coefficients for CO<sub>2</sub> dependence on CO were consistent throughout the lower part of the profile (200–1700 m), with overall mean value  $0.078 \pm 0.04$  (1 $\sigma$ ) ppm/ppb. This value lies within the range observed for biomass burning [Andreae et al., 1988] and large-scale mixing (0.04–0.1).

[37] Residual variance for O<sub>3</sub> increased with altitude as concentrations increased, in contrast to the results for CO<sub>2</sub>. Coefficients for CO in the O<sub>3</sub> fits were variable because the dependence of O<sub>3</sub> on CO was weak (Table 3). Ozone varied less than CO<sub>2</sub> with time-of-day but more from flight-to-flight. To test if flight-to-flight variance affected derived values of  $q_{ib}$  and  $q_{ih}$  for ozone, we constructed another linear model by adding a factor variable for each of the 15 flights. The model produced values of ( $O_{3b} - O_{3h}$ ) very similar to results both from the Linear Model and from conditional sampling, and we concluded that variability in background O<sub>3</sub> did not affect the analysis.

## 5. Results: Budget of CO<sub>2</sub> Over Amazonia

[38] Figures 6a–6d show profiles for CO<sub>2</sub> and O<sub>3</sub> from the Linear Model, with influences of combustion and large-

scale exchange removed by setting the concentration of CO in equation (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  $\pm 0.3$  ppm CO<sub>2</sub> or less.

[39] The Linear Model functions provide smooth vertical profiles for each hour, and smooth diurnal variations for each altitude (excepting 14 hours; see Figures 6a and 6b). Since each altitude and hour was treated independently, the smooth behavior supports the validity of the data averaging. Mean hourly and daily values of  $q_{ib}$  and  $q_{ih}$  are shown in Figures 6c–6f, derived from profiles in Figures 6a and 6b by averaging from 0–2500 m or 0–3300 m. Tower data for 41 m were adopted for 0–75 m, and values of  $q_{ib}$  and  $q_{ih}$  at 14 LT were replaced with the mean of 13 and 15 LT as noted above. It appears that CO<sub>2</sub> was added to the column after 17 hours, consistent with the flux data from the Manaus tower (Figure 4) and data from ABLE-2A [Wofsy et al., 1988], but data were too few to give reliable values for  $q_b$  at 1700 LT.

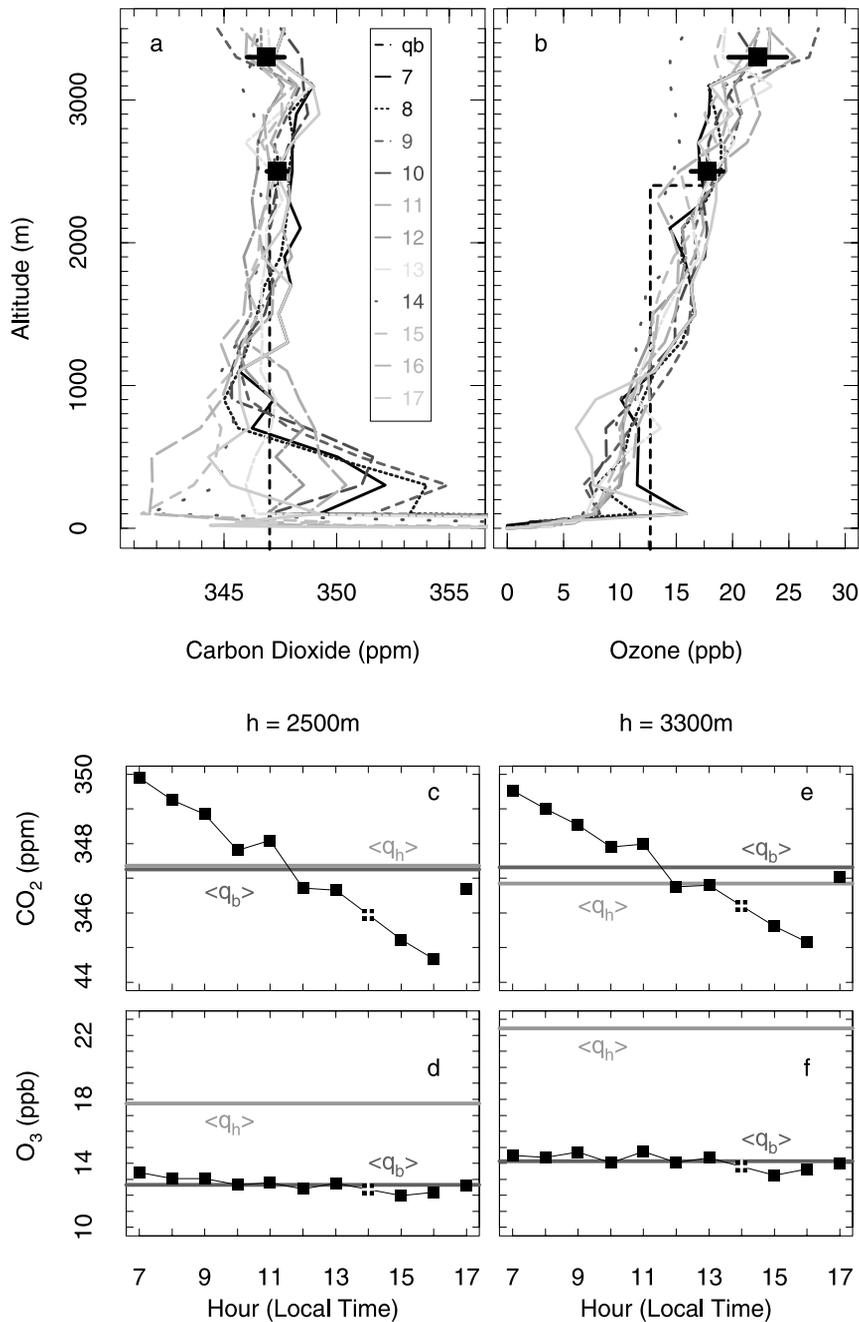
[40] We fit smooth curves to the profiles in Figures 6a and 6b and computed residuals, to estimate random errors at each altitude. The estimated uncertainties for CO<sub>2</sub> at each hour and altitude were smaller than  $\pm 1$  ppm. Uncertainties in mean column concentrations were estimated by fitting straight lines to the data in Figures 6c–6f and computing standard errors of the value at 12 LT; results were smaller than  $\pm 0.3$  ppm. Corresponding values for O<sub>3</sub> were  $\pm 2$  ppb and  $\pm 0.5$  ppb, respectively.

[41] Tables 4a (O<sub>3</sub>) and 4b (CO, CO<sub>2</sub>) give values for ( $q_{ib} - q_{ih}$ ) averaged over daytime (i.e., between the turning points, 8–17, covering hours 8–16 hours in Figure 6), which we will equate to  $\Delta q_i$ , for three treatments: “all data,” block averaged by altitude (no linear model or conditional sampling); Linear Model, equation (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<sub>bkgd</sub>,” equation (2) with CO set to

**Table 3.** O<sub>3</sub> Linear Model

Alt. (m)	Mean	$\sigma_r^a$	R <sup>2</sup>	$b_{j1}$
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

<sup>a</sup> $\sigma_r$ , square root of the residual variance (ppm for CO<sub>2</sub>, ppb for O<sub>3</sub>).



**Figure 6.** Concentrations for CO<sub>2</sub> and O<sub>3</sub> from Model 1, with CO set to 84 ppb and tower data appended at the bottom: (a) and (b) mean hourly profiles for CO<sub>2</sub> and O<sub>3</sub>, respectively; solid squares, daily mean at h = 2.5 and 3.3 km, vertical dashed line, q<sub>b</sub>; (c) and (d) for h = 2.5 km: solid squares, hourly column-means for CO<sub>2</sub> and O<sub>3</sub>; (light gray, q<sub>b</sub>), daily averages for 0-h; (dark gray, q<sub>h</sub>), at h; (e) and (f) same as Figures 6c and 6d, for h = 3.3 km. Hour = 14 was interpolated, and hour 17 was excluded from the computation of regional flux. See color version of this figure at back of this issue.

84 ppb. Results for “All Data” and Linear Model with CO<sub>obs</sub> are similar for both CO<sub>2</sub> and O<sub>3</sub> in most cases, indicating fidelity of the linear functions. The conditional sampling results were very close to the Linear Model with CO<sub>bkgd</sub>. In almost all cases, using observed CO gives a more positive value for Δq than CO<sub>bkgd</sub> for both CO<sub>2</sub> and O<sub>3</sub>, reflecting combustion-derived CO<sub>2</sub> (~0.6 ppm) and pollution-derived O<sub>3</sub>. These are relatively small corrections.

**Table 4a.**  $\overline{\Delta q_i} \equiv (q_b - q_h)$  Diurnal Mean for Different Data Treatments: ΔO<sub>3</sub><sup>a</sup>

Altitudes (m)	All data	Linear Mdl [CO <sub>obs</sub> ]	Linear Mdl [CO <sub>bkgd</sub> ]
0–3300	–5.40	–7.73	–8.24
30–2500	–4.48	–4.80	–5.05

<sup>a</sup>Units are ppbv.

**Table 4b.**  $\overline{\Delta q_i} \equiv (q_b - q_h)$  Diurnal Mean for Different Data Treatments: CO<sub>2</sub> and  $\Delta$ CO<sub>2</sub><sup>a</sup>

Treatment	$q_{h=3300}$	$q_b$ (0–3300)	$\Delta q_{3300}$	$q_{h=2500}$	$q_b$ (0–2500)	$\Delta q_{2500}$
“All data”	347.80	347.79	-0.01	347.45	347.85	0.40
Linear	346.97	347.80	0.83	347.51	347.84	0.33
Mdl <sub>[CO<sub>obs</sub>]</sub>						
Linear	346.85	347.10	0.26	347.36	347.02	-0.33
Mdl <sub>[CO<sub>bkgd</sub>]</sub>						

<sup>a</sup>Units are ppmv. “All data”: block averaged data by hour and 200 m altitude intervals (no linear model); “Linear Mdl”, equation (2) with CO = block-averaged (time/height) mean; “Linear Mdl, CO<sub>bkgd</sub>”, equation (2) with CO set to background (84 ppb).

[42] Despite the large diurnal variation in the lower part of the profile, results for the daytime average of  $((\text{CO}_2)_b - (\text{CO}_2)_h)$  were surprisingly close to zero for both values of  $h$ ,  $0.6 \pm 0.4$  ppm without compensating for CO, and  $-0.03 \pm 0.4$  ppm when the covariance with CO was removed. The estimated uncertainty for this result accounts for  $\pm 0.3$  ppm from the central values in Table 4b, plus the uncertainties for the average column amount (see discussion of Figure 6). An additional error of  $\pm 0.3$  ppm might result from diurnal bias, if our assignments of turning points were off by  $\pm 1/2$  hour.

[43] The null result for  $\overline{\Delta q_i} = 0 \pm 0.7$  ppm for CO<sub>2</sub> (with conservative uncertainty, i.e., adding random and possible bias errors) contrasts markedly with the depletion of O<sub>3</sub> in the column (0,  $h$ ),  $-6.4 \pm 2.5$  ppb (Table 4a). The result also contrasts with CO<sub>2</sub> data over the US in summer [Gerbig *et al.*, 2001], where the atmosphere clearly reflected the activity of surface vegetation. Over well-watered forests, the lower atmosphere was depleted by  $-5$  to  $-16$  ppm CO<sub>2</sub>, whereas in drought-impacted areas CO<sub>2</sub> was in excess by  $4$ – $10$  ppm. The US data support our concept that net sources or sinks of CO<sub>2</sub> should emerge as detectable contrasts between daily-average mean concentrations below  $h$  and above  $h$ . We conclude from the  $\sim 0$  contrast over Amazonia that the CO<sub>2</sub> budget was very close to balance in April 1987.

[44] Without tower data to define CO<sub>2</sub> 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], which was smaller than the uncertainty in PBL hourly values. Nevertheless it represents a potential source of systematic bias and should be accounted.

[45] Regional daytime CO<sub>2</sub> uptake at midday, computed from the slope of CO<sub>2</sub> column amounts, was  $-6.3 \mu\text{mol m}^{-2} \text{s}^{-1}$  for both  $h = 2500$  m and  $h = 3300$  m, slightly lower than daytime fluxes at Manaus (average  $-10.2 \mu\text{mol m}^{-2} \text{s}^{-1}$  [Fan *et al.*, 1990]). Tower uptake fluxes were  $\sim 30\%$  greater in the morning than the afternoon, due to more cloudiness and stomatal closure in the afternoon. Aircraft data were more symmetrical. The lower uptake rate and greater symmetry for aircraft data may both reflect the influence of wetlands, rivers, and inundated forest, which emit CO<sub>2</sub> at all hours. The tower fetch did not include significant areas of inundation or wetlands.

[46] The net 24-hour biotic exchange fluxes for CO<sub>2</sub> appear to be quite small,  $-0.13$  or  $+0.07 \mu\text{mol m}^{-2} \text{s}^{-1}$  for  $h = 2500$  or  $3300$  m respectively, using equation (1d) and results for O<sub>3</sub> (taking  $\Delta$ O<sub>3</sub> from Table 4a, O<sub>3</sub> flux from Fan

*et al.* [1990]). Daily mean uptake corresponding to an annual rate of only  $-0.25 \mu\text{mol m}^{-2} \text{s}^{-1}$  ( $1 \text{ ton C ha}^{-1} \text{ yr}^{-1}$ ) would correspond to  $\Delta$ CO<sub>2</sub> of  $-0.8$  ppm, at the extreme end of our error bounds for  $\Delta$ CO<sub>2</sub>, with additional allowance for uncertainty in the mean O<sub>3</sub> flux over Amazonia.

[47] Concentrations of CO were lower in ABLE-2B than in ABLE-2A [Sachse *et al.*, 1988]. If we attribute most of the CO enhancements over background to combustion, the associated CO<sub>2</sub> flux from the region was only  $+0.25 \mu\text{mol m}^{-2} \text{s}^{-1}$  (comparing  $\Delta$ CO<sub>2</sub> for CO<sub>obs</sub> versus CO<sub>bkgd</sub>). These low rates of biomass burning in the wet season nevertheless exceeded regional biogenic uptake.

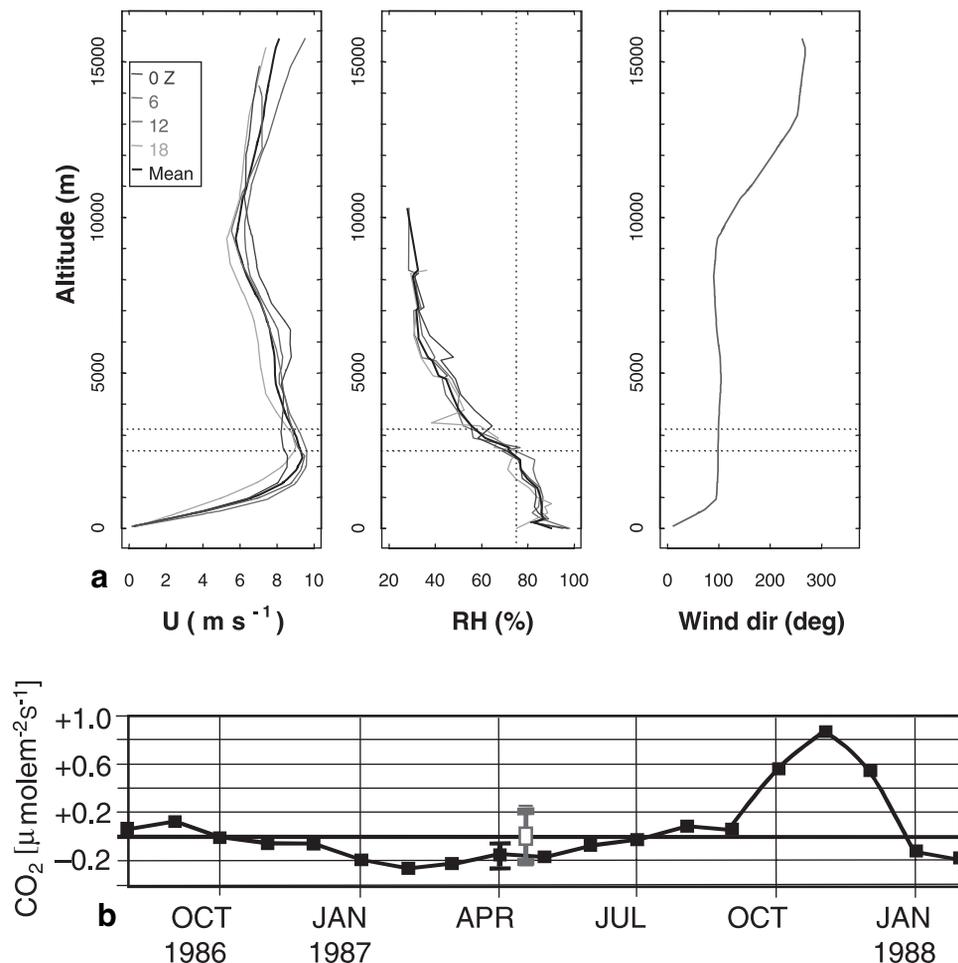
[48] We estimated the fetch for the aircraft measurements using ozone data combined with rawinsonde measurements, summarized in Figure 7a. Six stations made four soundings daily in and around the Basin. The density-weighted mean wind for  $0$ – $3.3$  km averaged  $6.4 \text{ m s}^{-1}$  from the ESE in the main area of aircraft operations (station EMBRAPA), with similar values upwind at Belém and Alta Floresta. The coast is about  $\sim 2400$  km ESE of our operational site, giving an average advection time,  $t_{\text{adv}}$ , of 4.3 days. From the O<sub>3</sub> profiles, which satisfied the quasi-periodic condition, we infer  $\tau_{\text{exch}} = 2.7$  days from equation (1c) and Table 4a. Pereira [quoted by Jacob and Wofsy, 1990] measured <sup>222</sup>Rn on the Electra, and inferred  $\tau_{\text{exch}} \sim 3$  days for the PBL, in harmony with our value. The observation that the advection time exceeds  $\tau_{\text{exch}}$  lends support to the analysis. Likely the true residence time in the Basin, for air in our primary study area, is much longer than  $t_{\text{adv}}$  due to variance of the wind associated with rain storms, river breeze, inhomogeneous surface heat fluxes, etc. This would further reduce errors associated with our approximate treatment of transport.

[49] The values of  $t_{\text{adv}}$  and  $\tau_{\text{exch}}$  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, and mean residence times may be lengthened by dispersive processes. The width of the fetch is probably comparable to the meridional extent of the principal convergence zone in Amazonia, roughly  $\pm 500$  km. It seems clear that the aircraft observed a vast region of mostly intact equatorial forest and associated mosaics of wetlands, inundated lands (varzea, igapo), as well as the great rivers ( $20$ – $30\%$  of the land area) at a time of high, and increasing, water levels.

## 6. Discussion

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

[51] Model results (Figure 7b; Botta and Foley [2001]) indicate that forest growth should have been near a seasonal



**Figure 7.** (a) Wind speed ( $\text{m s}^{-1}$ ), relative humidity (%), and wind direction (degrees) at EMBRAPA (60 W, 2.5 S): Averages of four soundings per day (0, 6, 12, 18 GMT, or 20, 2, 8, and 14 local) obtained daily during the experiment [Cohen *et al.*, 1995]; dotted lines show  $h = 2500$  and  $3300$  m. The sounding data for 5 sites may be retrieved from <ftp://ftp-gte.larc.nasa.gov/pub/ABLE2B/GROUND/NOBRE.INPE/>. (b) Ecosystem flux for April–May, 1987, computed by the IBIS model (gray symbol) for the study area (58–60W, 1–3S), compared to the regional flux results from ABL2B (black symbol) (IBIS results courtesy A. Botta and J. Foley, private communication, 2001). Note the ENSO-induced release of carbon in Oct.–Dec. 1987, with strong uptake in the previous wet season (Jan.–May). See color version of this figure at back of this issue.

high in April, 1987. The model computed net exchange of CO<sub>2</sub> for Amazonia 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 7b 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 sunnier, drier conditions than average. Models indicated release of CO<sub>2</sub> due to drought later in 1987 [cf. Tian *et al.*, 1998], well after ABL2B.

[52] Our observations do not show the uptake indicated by the model, although the uncertainty ranges stretch sufficiently to overlap the range of model fluxes in central

Amazonia (Figure 7b). Since deforestation and biomass burning are both at seasonal lows late in the wet season, and drought had not yet begun, we expected significant net uptake. Recent tower results from Santarém showed release of CO<sub>2</sub> from an old-growth equatorial forest in the wet season, and uptake in the dry season [Saleska *et al.*, 2001], consistent with the central values from the ABL2B aircraft data.

[53] If confirmed, these results would suggest that seasonal variations of respiration may be more important than considered hitherto. Rivers and flooded lands are large, persistent sources of CO<sub>2</sub> to the atmosphere [Wofsy *et al.*, 1988; Richey *et al.*, 1990], and a significant quantity of this CO<sub>2</sub> arises from organic matter transported to watercourses from *terra firma* forests and marginal wetlands [Quay *et al.*, 1992]. Tower studies and ecological models generally focus on forests in upland areas, whereas aircraft data integrate over the landscape. Thus we might 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. All stimulate decomposition. A plausible explanation for lack of CO<sub>2</sub> uptake in the wet season is that forest growth is masked by seasonal enhancement of decomposition. To test this hypothesis, seasonal variations of both growth and decomposition should be examined, as well as linkages between components of the landscape mosaic.

## 7. Conclusions

[54] ABLE-2 provided the first comprehensive look at temporal and spatial distributions of CO<sub>2</sub> and other tracers in the tropical atmosphere. We analyzed the historical data for CO<sub>2</sub> in ABLE-2B to show that regional fluxes for CO<sub>2</sub> lead to atmospheric concentration gradients below 3 km that can be quantified by systematic aircraft soundings. A first-order assessment of regional net fluxes for April 1987 was derived by complementing aircraft data with tracer and meteorological data, and with measurements from flux towers. The analysis introduced a framework with (1) “air-mass following” analysis approach; (2) column budgets, instead of conventional CBL budgets; (3) multiple tracers (CO<sub>2</sub>, CO, O<sub>3</sub>); (4) tracer similarity analyzed by separation of timescales for the surface forcing; and (5) combination of tower data with the above to derive regional fluxes. Future improvements would allow more rigorous application of the column budget approach. Better characterization of fluxes from different vegetation types/ecosystems would be needed, along with application of state-of-the-art mesoscale transport models in place of the simple transport concepts available for ABLE-2B.

[55] Significant covariance was observed between CO<sub>2</sub> 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 CO<sub>2</sub> 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 O<sub>3</sub> 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 CO<sub>2</sub> could be estimated by similarity with O<sub>3</sub>.

[56] The layer-mean (0 to  $h = 3300$  m or 2500 m) concentration of CO<sub>2</sub> declined by 3–4 ppm from morning to evening, but the 24-hour average almost exactly equaled the value just above  $h$ . We infer that rates for daytime uptake were large, but there was no observable *net* uptake or release of CO<sub>2</sub> over 24 hours. The east-west fetch for these measurements, ~1500 km, encompassed a large fraction of the equatorial Amazon Basin. The lack of regional uptake for CO<sub>2</sub> 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 contrasted sharply with aircraft flights over the northern US in August, 2000.

[57] Our analysis draws attention to a potentially critical difference between studies at ecosystem scales, such as flux

towers and ecosystem models, versus regional scales. In Amazonia, up to 30% of the land surface may be inundated late in the wet season, providing strong sources of atmospheric CO<sub>2</sub>. Much of the carbon released from these areas is derived from terrestrial sources. Hence the regional CO<sub>2</sub> flux from Amazonia to the global atmosphere may be quite different than the exchange associated with an individual component, even a major component such as *terra firma* 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.

## Appendix A

[58] We derive a solution to equation (1b) for a case with surface fluxes consisting of two components: (1) a periodic term,  $S'_i$ , assumed to be invariant in the fetch and to have period 1 day ( $\equiv T$ ) and zero mean; and (2) a 24-hour net exchange, approximated as constant in the fetch. Thus  $S_i = \bar{S}_i + S'_i = \bar{S}_i + \sum_k \text{Re}[S_k \exp(ik\omega t)]$ , where  $\omega = 2\pi/T$  and  $T = 24$  hours. These assumptions allow us to express  $\Delta q_i$ , the difference between the column-mean concentration of species  $i$  between 0 and  $h$  and value at  $q_h$ , as the sum of a periodic part  $\Delta q'_i$  plus a non-periodic part,  $\Delta q_{ib}$ ,

$$\Delta q_i = \left(1 - e^{-t/\tau}\right) \bar{S}_i / (n_b h) + \sum_k \text{Re}[B_k \exp(ik\omega t)]. \quad (\text{A1})$$

where

$$B_k = S_k \frac{\tau[1 - ik\omega\tau]}{n_b h [1 + [k\omega\tau]^2]} = S_k \frac{T}{2\pi k n_b h} \left(-i + \frac{T/\tau}{2\pi k}\right).$$

Here we have assumed that  $\Delta q_i = 0$  at the initial time when air enters the Basin, and also that we observe air representing a random ensemble of entry times over 24 hours.

[59] The average value of  $\Delta q_i$  over 24 hours,

$$\frac{1}{T} \int_0^{t+T} \Delta q_i dt \equiv \overline{\Delta q_i}$$

is then given by

$$\overline{\Delta q_i} = \bar{S}_i \frac{\tau^2}{T n_b h} \left(e^{-t/\tau} - e^{-(t+T)/\tau}\right) \approx \bar{S}_i \frac{\tau}{n_b h} \left(1 - e^{-t/\tau}\right), \quad (\text{A2})$$

where the approximation holds for  $T/\tau \ll 1$  (i.e., exchange time longer than 1 day). We used the fact that all periodic terms vanish when averaged over 24 hours.

[60] There could be coupling between the diurnally varying transport and the slowly varying part of the column enhancement  $\Delta q_i$ , if the inverse exchange time  $1/\tau$  had a periodic variation that correlated with that of  $\overline{\Delta q_i}$ . Physically this means, that if convection occurred preferentially at times of day when concentrations of CO<sub>2</sub> in the PBL differed from the 24-hour mean value, transport through

level  $h$  could be biased and a non-zero value could develop for  $\overline{\Delta q_i}$  even if the 24-hour mean surface flux were zero (“rectification”). Fortunately, the distribution of convective rainfall over the day was almost symmetrical relative to the diurnal cycle of CO<sub>2</sub>: 45% of rainfall in the Basin occurred between midnight and 1300 LT, when concentrations CO<sub>2</sub> in the lower atmosphere exceed the 24-hour mean, with 55% at times when CO<sub>2</sub> was depleted by photosynthesis in the afternoon (Figure 5b; Greco *et al.* [1990]). Hence deep convection transported air aloft with mean CO<sub>2</sub> very similar to the 24-h mean, with at most a very small bias (10%) towards preferential export of lower concentrations.

[61] The forest takes up CO<sub>2</sub> during the daytime and releases it at night. Our daytime data measures the mass balance of the layer from 0 to  $h$ , giving the mean daytime surface flux in terms of the daytime mean of the time derivative of  $\Delta q_i$

$$\begin{aligned} \text{RE} \left[ \frac{2}{T} \int_{T/4}^{3T/4} \sum \frac{S_k}{n_b h} e^{2\pi i k t / T} dt \right] &= 0, \quad k \text{ even} \\ &= \text{RE} \left[ \sum \frac{2}{\pi k} \frac{S_k}{n_b h} i^{k-1} \right], \quad k \text{ odd} \end{aligned} \quad (\text{A3a})$$

where the integration limits go from sunrise ( $T/4$ ) to sunset ( $3T/4$ ). There is a small contribution to the daytime budget associated with non-periodic exchange through altitude  $h$ ,

$$\begin{aligned} \text{RE} \left[ \frac{2}{T} \int_{T/4}^{3T/4} \sum \frac{\partial \Delta q_i}{\partial t} e^{2\pi i k t / T} dt \right] &= 0, \quad k \text{ even} \\ &= \text{RE} \left[ \sum \frac{2}{\pi k} \frac{S_k}{n_b h} \left( i^{k-1} + i^k \frac{T/\tau}{2\pi k} \right) \right], \quad k \text{ odd.} \end{aligned} \quad (\text{A3b})$$

Note that the slowly varying exchange term does not contribute at all if  $S_k$  is real, i.e., if all source terms change sign at sunrise and sunset: if the system is close to a periodic steady state,  $\Delta q_i$  is higher than the steady-state value in the morning, and exchange with air aloft provides a flux correspondingly higher. However the value of  $\Delta q_i$  and associated exchange fluxes are lower by the same amount in the afternoon, canceling the morning effect and ensuring that flux associated with replacement of the mixed layer has no effect on the 12-hour mean value for  $\Delta q_i$ . Since fluxes of CO<sub>2</sub> from vegetation approximate real, even periodic functions, the value derived for daytime uptake from  $\partial \Delta q_i / \partial t$  is insensitive to the slow process of PBL exchange.

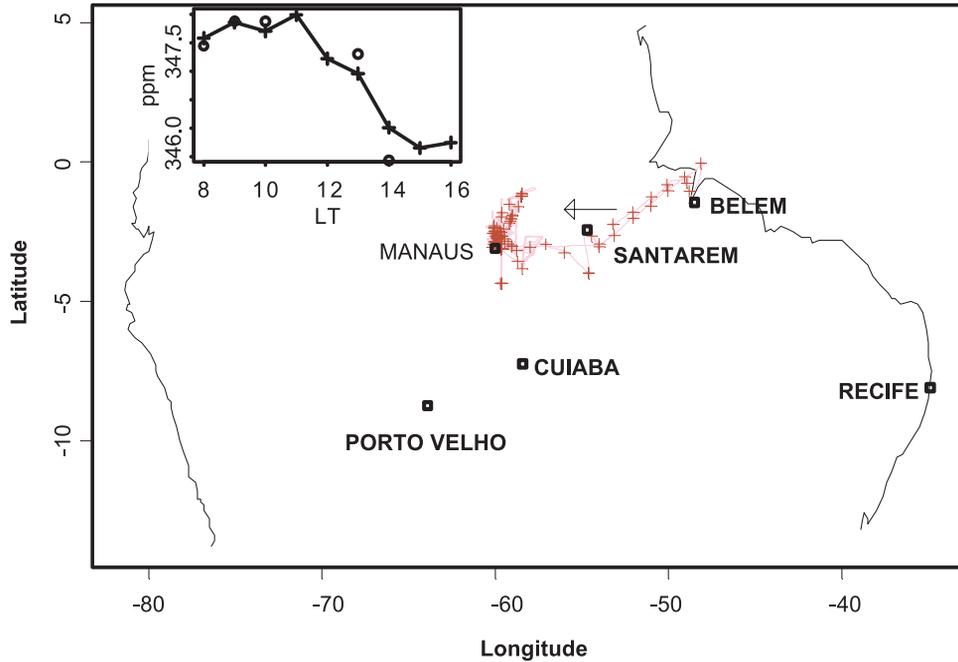
[62] **Acknowledgments.** We gratefully acknowledge the contributions of the staff of the Global Tropospheric Experiment and NASA’s Langley Research Center and the pilots and crew from the Wallops Flight Facility, who made this work possible. A. D. Botta and J. Foley (University of Wisconsin) generously provided unpublished results from their Amazon model study. This work was supported by grants to Harvard University for airborne CO<sub>2</sub> studies (NASA NAG2-1310) and the Large-Scale Biosphere-Atmosphere study in Brazil (NASA NCC5-341) and by funding for the CO<sub>2</sub> Budget and Rectification Airborne Study (Department of Energy, DOE DE-FG02-98ER62695, the National Science Foundation, NSF ATM-9821044).

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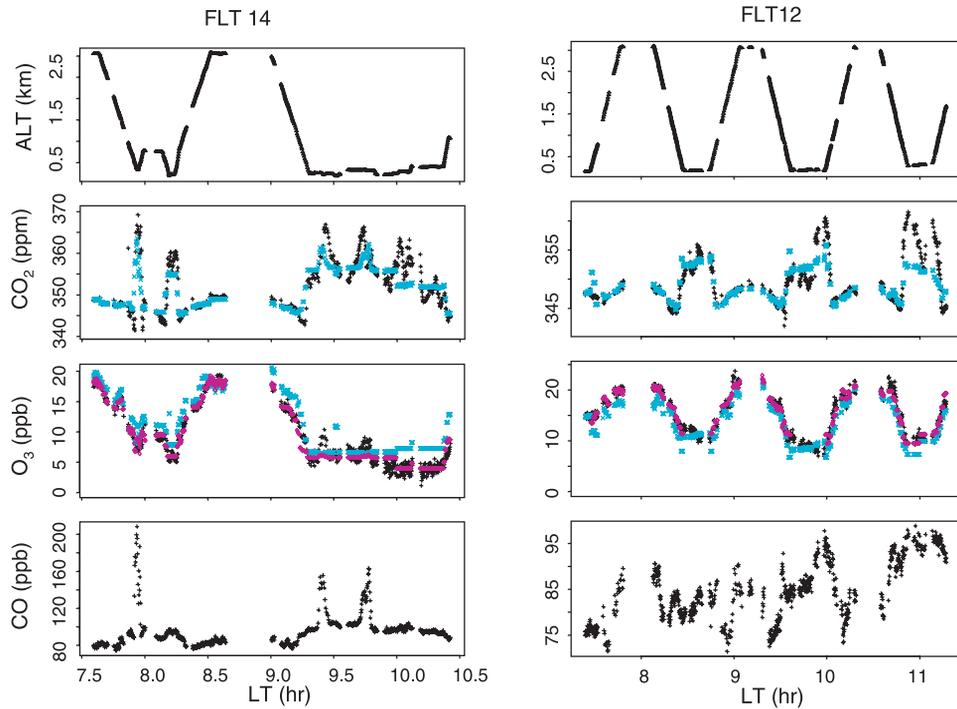
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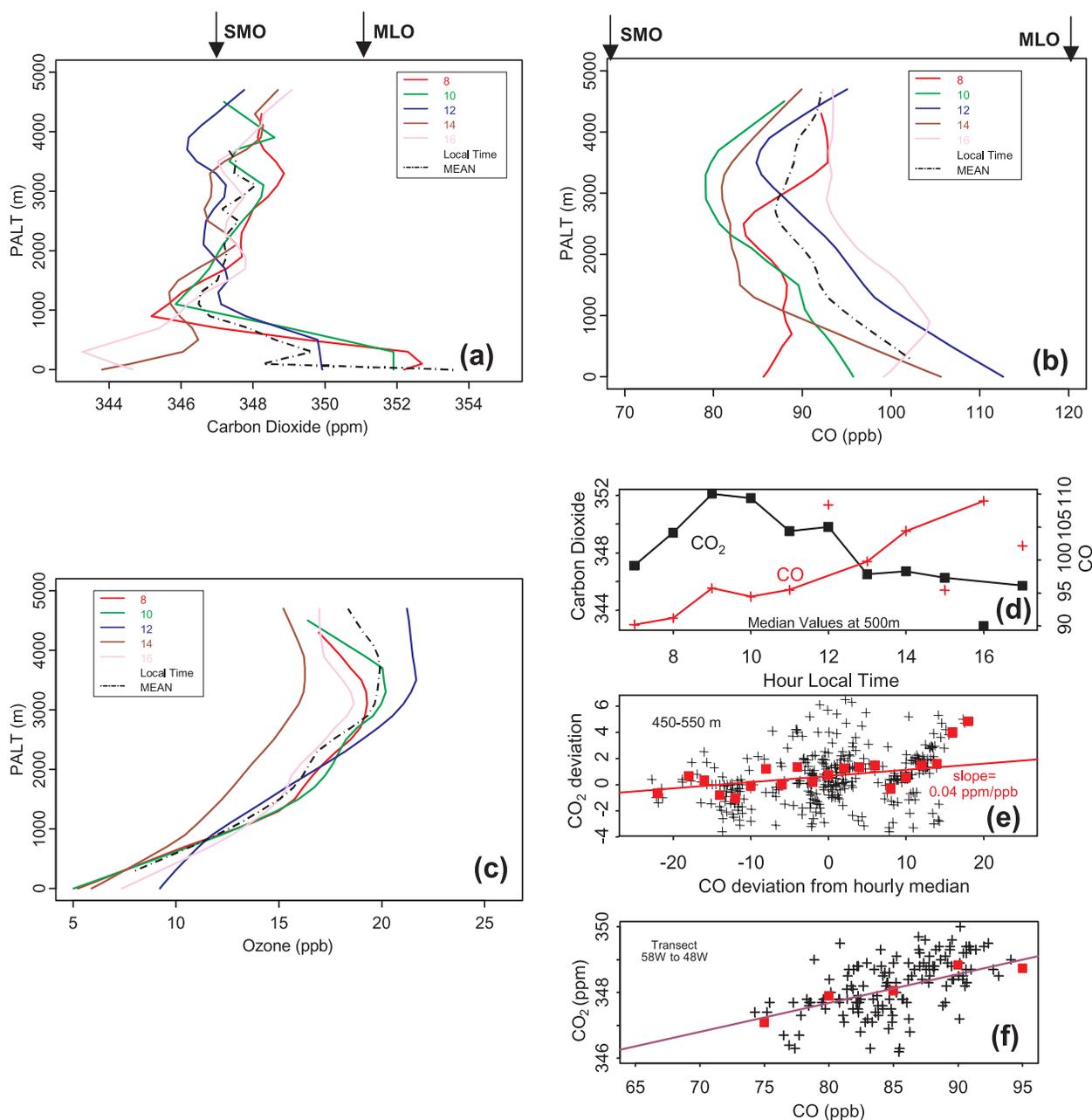
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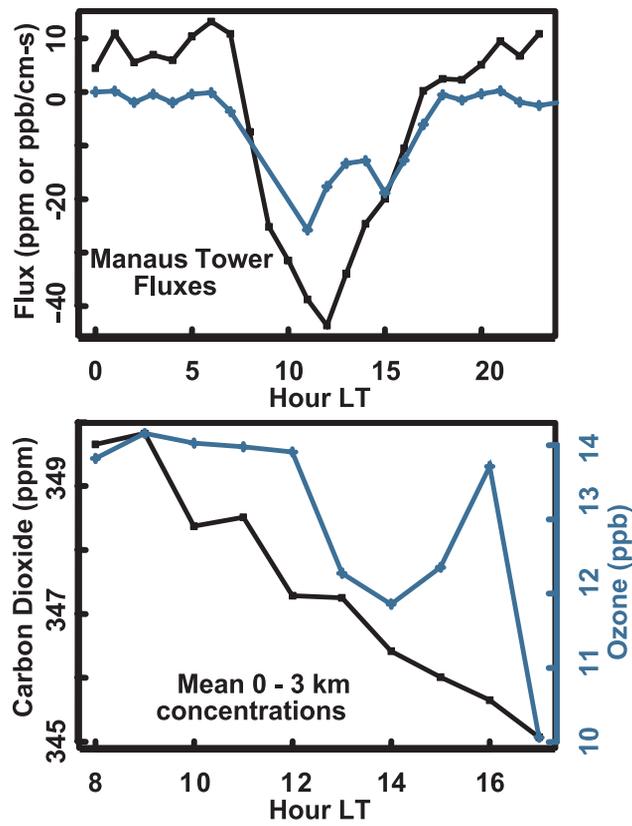
**Figure 1.** Map showing locations (pluses) for the 85 vertical profiles acquired during ABLE-2B,15 April –8 May 1987. The wind in the lower 3 km of the atmosphere averaged  $6.4 \text{ m s}^{-1}$  from the East (arrow), giving a transit time of 4.3 days to the main sampling area northeast of Manaus, 2400 km from the ocean. (inset) Diurnal variation of the mean column concentration of CO<sub>2</sub> (0.2–2.8 km) in central Amazonia (pluses, west of  $-56^\circ$ ) compared to values in eastern Amazonia (circles, east of  $-56^\circ$ ).



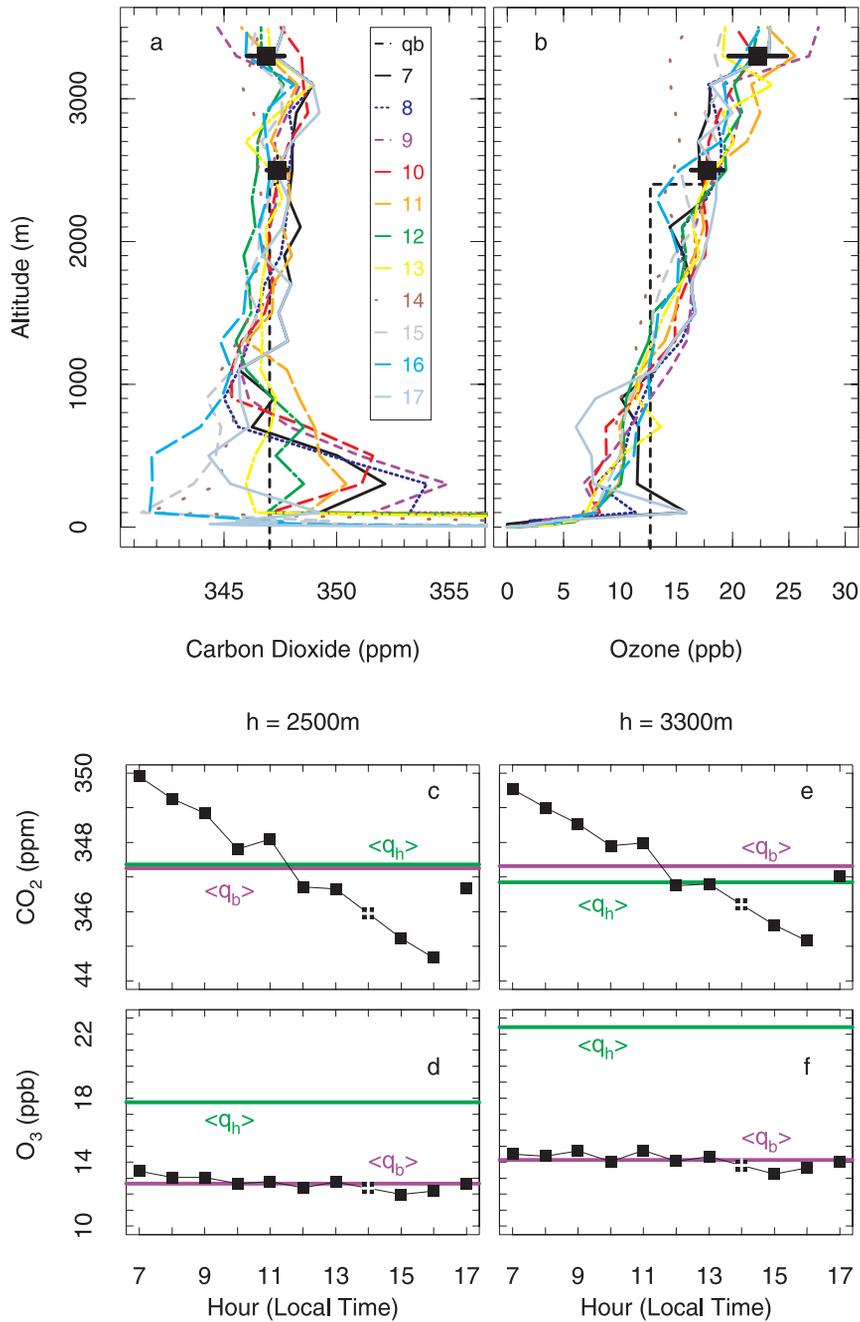
**Figure 2a.** Data for altitude, CO<sub>2</sub>, O<sub>3</sub>, and CO are plotted versus time for flights 12 and 14 (see Table 1). Black points show the observations for 10 s intervals with valid data for CO<sub>2</sub>. Blue points show linear model representation of the data using CO as a predictor with hour and altitude as factors. Red points for O<sub>3</sub> add date as a factor to account for variation of background O<sub>3</sub> concentrations. Smoke layers encountered on flight 14 (8, 9.5, 9.7 hours) contributed up to 15 ppm CO<sub>2</sub>.



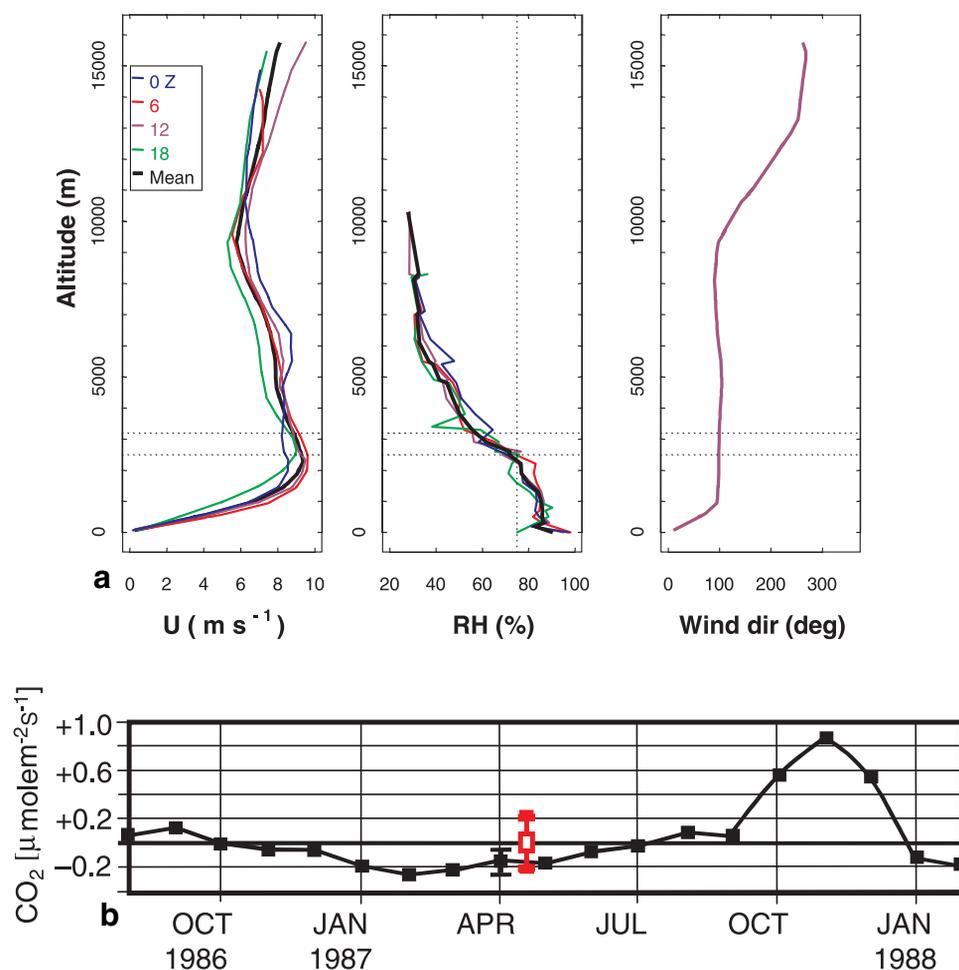
**Figure 3.** (a) Vertical profiles for CO<sub>2</sub> from 15 flights, days 105–126, 1987, in central Amazonia (Table 1), block averaged by local time. (b) Vertical profiles for CO, as in Figure 3a. Arrows show data from mid-Pacific stations Samoa (SMO, 14° 15'S, 170° 34'W) and Mauna Loa (MLO, 19° 32'N 155° 35'W) for April 1987 (CO<sub>2</sub>) and for April 1990 (the first year of station data for CO). (c) Vertical profiles for O<sub>3</sub>, as in Figure 3a. (d) Diurnal variation of CO<sub>2</sub> and CO concentrations at 500 m (median values for each hour, all flights). Lines illustrate the general trends during the day. (e) Deviations of CO<sub>2</sub> and CO from the hourly median concentrations (Figure 3d) at 500 ± 50 m. (f) CO and CO<sub>2</sub> gradients across the Amazon Basin (Santarém-Manaus survey) at 3 km altitude: pluses, raw; minuses, block averaged by CO (1 ppb bins, Figure 3e; 5 ppb bins, Figure 3f), linear regression line; straight-line connecting SMO and MLO data points (slope = 0.08 ppm/ppb).



**Figure 4.** (top) Fluxes of CO<sub>2</sub> (black) and O<sub>3</sub> (gray) versus local time at the Manaus tower [Fan *et al.*, 1990]. (bottom) Density-weighted mean concentrations during the day for the column 0–3.1 km from the aircraft.



**Figure 6.** Concentrations for CO<sub>2</sub> and O<sub>3</sub> from Model 1, with CO set to 84 ppb and tower data appended at the bottom: (a) and (b) mean hourly profiles for CO<sub>2</sub> and O<sub>3</sub>, respectively; solid squares, daily mean at h = 2.5 and 3.3 km, vertical dashed line, q<sub>b</sub>; (c) and (d) for h = 2.5 km: solid squares, hourly column-means for CO<sub>2</sub> and O<sub>3</sub>; (light gray, q<sub>b</sub>), daily averages for 0-h; (dark gray, q<sub>h</sub>), at h; (e) and (f) same as Figures 6c and 6d, for h = 3.3 km. Hour = 14 was interpolated, and hour 17 was excluded from the computation of regional flux.



**Figure 7.** (a) Wind speed ( $\text{m s}^{-1}$ ), relative humidity (%), and wind direction (degrees) at EMBRAPA (60 W, 2.5 S): Averages of four soundings per day (0, 6, 12, 18 GMT, or 20, 2, 8, and 14 local) obtained daily during the experiment [Cohen *et al.*, 1995]; dotted lines show  $h = 2500$  and  $3300$  m. The sounding data for 5 sites may be retrieved from <ftp://ftp-gte.larc.nasa.gov/pub/ABLE2B/GROUND/NOBRE.INPE/>. (b) Ecosystem flux for April–May, 1987, computed by the IBIS model (gray symbol) for the study area (58–60W, 1–3S), compared to the regional flux results from ABL2B (black symbol) (IBIS results courtesy A. Botta and J. Foley, private communication, 2001). Note the ENSO-induced release of carbon in Oct.–Dec. 1987, with strong uptake in the previous wet season (Jan.–May).