The CO₂ Budget and Rectification Airborne Study: Strategies for Measuring Rectifiers and Regional Fluxes

Britton B. Stephens¹, Steven C. Wofsy², Ralph F. Keeling¹, Pieter P. Tans³, and Mark J. Potosnak⁴

Temporal covariations between atmospheric transport and surface gas fluxes, known as rectifier effects, may lead to significant spatial variations in CO₂ and other gases. Attempts to constrain carbon fluxes using surface background measurements and inverse methods are limited by large uncertainties in these effects. Airborne measurements of the vertical and horizontal distributions of CO₂, CO, O₂, and other tracers have the potential to significantly reduce these uncertainties. In this paper, we discuss observational requirements for quantifying rectifier effects and estimating regional fluxes, and use existing temperate and tropical forest data and model simulations to investigate the most promising measurement strategies. Using relationships between multiple tracers, it should be possible to separate the terrestrial, oceanic, and industrial influences on observed horizontal and vertical CO₂ variations. Summer observations of terrestrial CO₂ signals over land would then provide a direct measure of the diurnal rectifier effect, while comparisons between summer and winter observations would constrain the seasonal rectifier. We have plans for such measurements over North America and the adjacent ocean regions using airborne instrumentation and flask samples, as part of the CO₂ Budget and Rectification Airborne Study. The analyses presented here provide preliminary tests of our measurement approach and reveal evidence of strong diurnal rectifier effects.

1. INTRODUCTION

The sources of atmospheric carbon dioxide (CO₂), including fossil fuel combustion and biomass burning, appear to exceed the sum of atmospheric build up and oceanic uptake by 2 billion tons of carbon per year [e.g. Schimel et al., 1995]. To understand the response of global ecosystems to anthropogenic perturbations and to predict future trends in atmospheric CO₂, it is critical to determine where this excess carbon is being stored and to define the mechanisms for sequestration. Unfortunately, spatial and temporal heterogeneities in ocean and land biospheric CO₂ fluxes make it impractical to derive regional or global values from direct measurement at small scales. Instead, oceanic and terrestrial CO₂ fluxes are currently estimated by inverting measurements of atmospheric CO₂ [Enting and Mansbridge, 1989; Fan et al., 1998; Baker, 1999; Dargaville and Simmonds, 1999; Taguchi, 1999], CO₂ and \(^{13}\)CO₂/\(^{12}\)CO₂ [Enting et al., 1993; Ciais et al., 1995], CO₂ and C\(^{18}\)O\(^{16}\)O/C\(^{16}\)O\(^{16}\)O [Peylin et al., 1999], or CO₂
and O₂/N₂ [Keeling et al., 1996] at background stations using numerical transport models.

The background surface stations are located, and sampling times selected, to obtain data representing the largest spatial scales in order to facilitate the determination of global trends. Consequently, most stations are remote from strong source or sink regions and measurement protocols stress sampling of air uncontaminated by regional surface processes. Since the network design deliberately minimizes the influence of surface exchange, the station data are not ideal for determining the loci and magnitudes of regional fluxes, or for understanding gradients associated with atmosphere-surface exchange. It is therefore not surprising that estimates of regional CO₂ sources and sinks derived from inverse model calculations are model-dependent, and especially sensitive to model representations of vertical mixing and horizontal advection. Additional measurements are needed at altitude and over the continents to reduce these uncertainties.

2. RECTIFIER EFFECTS

Denning et al. [1995] showed that temporal covariations between terrestrial CO₂ fluxes and rates of vertical mixing through the planetary boundary layer (PBL) may produce vertical and horizontal CO₂ gradients as large or larger than those resulting from net industrial, terrestrial, or oceanic exchanges. Over the continents during summer, vertical mixing through the PBL is vigorous and the PBL is relatively deep. There is a deficit in CO₂ near the surface associated with the net seasonal flux of CO₂ into the terrestrial biota, but the magnitude of this deficit is mitigated by dilution in the deep PBL [e.g. Bakwin et al., 1998]. In contrast, during winter the PBL is shallow and the increase in CO₂ resulting from the net terrestrial efflux is trapped near the surface. Thus seasonal terrestrial CO₂ exchange, even with zero net annual flux, produces higher annual mean CO₂ concentrations at the surface over land, and model simulations indicate that this elevation of annual-mean concentration propagates over the oceans to remote stations [Denning et al., 1996].

This and other similar processes have been termed “rectifier” effects, by analogy to an electronic circuit that converts an alternating current with a mean of zero volts to a direct current with a finite mean voltage. To begin, we define a rectifier effect as any temporal covariation between a surface flux and atmospheric mixing or transport that produces a time-mean spatial concentration gradient in the atmosphere. This broad definition includes a) the vertical-seasonal terrestrial-CO₂ rectifier just described, b) a similar vertical-diurnal terrestrial-CO₂ rectifier, c) the horizontal-seasonal terrestrial-CO₂ effect described by Pearman and Hysom [1980], d) O₂ and isotope-ratio rectifiers corresponding to these terrestrial-CO₂ effects, and e) the horizontal-seasonal and vertical-seasonal marine-O₂ rectifiers described by Stephens et al. [1998] and Stephens [1999]. Here, we are only concerned with the vertical-seasonal and vertical-diurnal terrestrial-CO₂ rectifier effects, which we will refer to in the remainder of the paper simply as the seasonal and diurnal rectifiers.

The diurnal rectifier, which operates primarily during summer, is depicted in Figure 1. In daytime the photosynthetic uptake is distributed through a thick atmospheric layer associated with the deep PBL. At night the PBL collapses, holding the respired flux near the surface through the night and morning and stranding the low concentrations aloft. Thus over much of the day, and on average, concentrations are higher near the surface over land than at upper levels and over the oceans, even in the presence of net continental CO₂ uptake. Using only surface concentration measurements, it is not possible to distinguish such time-mean signals from those resulting from a net terrestrial CO₂ source.

The model of Denning et al. [1996] indicates that the seasonal rectifier accounts for 80% of the total rectification effect, of approximately 2 ppm, predicted at northern marine boundary-layer stations. However, greater uncertainty may be associated with the influence of the diurnal rectifier as it produces even higher predicted concentration anomalies at the surface over the continents, of up to 18 ppm [Denning et al., 1996], and its effect on the marine boundary-layer stations depends critically on the complex coupling between mixing over the continents and over the oceans. If natural rectifiers produce an interhemispheric CO₂ difference on the order of 2 ppm, as several of the models in the TRANSCOM study [Law et al., 1996] predict, an inversion calculation using a model without such rectification would indicate a large, spurious net CO₂ source in the Northern Hemisphere, and vice versa.

Figure 2 shows seven-day averages of CO₂ versus height and longitude at 43°N predicted by the TM2 atmospheric transport model [Heimann, 1995], for combinations of realistic oceanic, terrestrial, and fossil fuel sources [Keeling et al., 1989] during winter and summer. The longitudinal patterns in Figure 2 reflect vertical mixing of CO₂ as air moves eastward over North America. The net effect of the terrestrial biosphere is to make North America a source for CO₂ during win-
ter and a sink during summer. Because TM2 lacks a diurnal rectifier, it predicts a drawdown of CO$_2$ near the continental surface during summer (Figure 2b). As shown below, existing continental measurements actually show enhanced CO$_2$ concentrations in this region at this time.

While this discrepancy imparts considerable uncertainty to inverse calculations using such a model, inversions based on current models with more complex boundary-layer parameterizations are not necessarily more accurate. For example, an inverse calculation based on the Denning et al. [1996] model, which compared well with diurnal and seasonal continental surface observations, was unable to produce a realistic CO$_2$ sink distribution [Denning et al., 1999]. High-resolution airborne measurements of CO$_2$ and other tracers are needed to quantify the effect of rectifiers on surface background stations and to test the representation of vertical mixing in atmospheric transport models. Such measurements would also provide the opportunity to investigate methods of measuring surface CO$_2$ fluxes on regional and continental scales.

3. MEASUREMENT REQUIREMENTS

In order to quantify vertical and horizontal CO$_2$ gradients associated with the diurnal and seasonal rectifiers, and to provide data that can be directly compared to coupled terrestrial-atmosphere models, it is necessary to correct for oceanic and industrial influences. As discussed below, a fossil-fuel correction should be possible using correlations between CO$_2$ and O$_2$ [Bakwin et al., 1998], and between CO$_2$ and O$_2$, SF$_6$ and additional industrial tracers. Correlations between CO$_2$ and O$_2$ could also help to identify gradients resulting from air-sea gas exchange. Airborne measurements of other species and isotope ratios, such as CH$_4$, N$_2$O, O$_3$, $^{13}$CO$_2$/$^{12}$CO$_2$, and $^{18}$O$^{16}$O/$^{16}$O$_2$ would provide additional information on the influences of terrestrial exchange and boundary-layer mixing of observed CO$_2$ gradients, as well as valuable insights into their own source/sink distributions. Weekly airborne flask measurements of many of these species are currently underway. However, adequately resolving the horizontal and vertical propagation of terrestrial CO$_2$ signals through the boundary layer, which is ultimately responsible for rectification, requires measurements at significantly higher spatial and temporal resolution.

Intensive measurements at several different times of year would be required on repeated vertical profiles, covering multiple times of day and extending over a wide range of longitude. At a minimum, CO$_2$ and the predominately industrial tracer CO would need to be measured using continuous instrumentation. Ideally, such high-resolution measurements for CO$_2$ would produce comparable cross sections to Figures 2a and 2b. Using CO measurements to quantify the fossil fuel component, and to separate it from the terrestrial component of CO$_2$, would allow direct tests of model simulations of these two components (Figures 2c-f). Summer-time measurements of the magnitude of CO$_2$ enhancement in the PBL would provide direct tests of the diurnal rectification predicted by current and future models. Furthermore, it is the seasonal difference in the efficiency of mixing through the PBL that drives the seasonal rectifier. This mixing controls the rate of vertical propagation, with longitude, of the continental signals shown in Figure 2. Therefore, airborne measurements of the propagation of these signals during summer and winter should provide quantitative tests for model representations of the seasonal rectifier.

In regions remote from oceanic influence, it should be possible to verify the correction for fossil fuel using flask-sample O$_2$ measurements and differences in the oxidation quotient. Terrestrial photosynthesis and respiration exchange O$_2$ and CO$_2$ in the ratio 1.11:0.5 [Severinghaus, 1995], whereas non-coal fossil fuel oxidation ratios average approximately 1.4:1.0 [Keeling, 1988]. Measurement of O$_2$:CO$_2$ and O$_2$:CO ratios would therefore provide strong checks on the separation of terrestrial and industrial signals, and help to identify anomalies or errors. Flask measurements of SF$_6$ could provide additional tests for the identification of industrial
signals using continuous CO data. Measurements of \(^{14}\text{CO}_2/^{12}\text{CO}_2\) ratios offer the most robust constraint on industrial \(\text{CO}_2\) signals, as fossil fuels have virtually no \(^{14}\text{C}\), however the sample-volume requirements for these measurements are somewhat more demanding than for other tracers.

In an attempt to quantify terrestrial influences on observed \(\text{CO}_2\) gradients, flask-sample \(\text{O}_2\) measurements could also play an important role in identifying regions affected by air-sea exchange. It is possible to define a derived tracer, atmospheric potential oxygen (APO ≈ \(\text{O}_2 + 1.1 \text{ CO}_2\)), that is conserved with respect to terrestrial biotic exchange [Stephens et al., 1998]. After removing fossil fuel influences on observed \(\text{CO}_2\) and \(\text{O}_2\) gradients using CO correlations, variations in APO should reflect only oceanic exchange. Therefore, air-
borne observations of APO gradients over continents would indicate the potential presence of oceanic CO₂ signals. If estimates of the marine O₂:CO₂ flux ratio were also available from a biological ocean model, then these APO gradients could provide estimates of the direct influence of air-sea exchange on the CO₂ measurements. Airborne measurements of radon would also help to identify marine air that had recently been influenced by terrestrial exchange, and to quantify vertical mixing rates over the continents [Ramonet et al., 1996].

Observations of the propagation of oceanic signals over land and terrestrial signals over the oceans would provide a basis for investigating the coupling between terrestrial and oceanic boundary-layer mixing, helping to determine the impact of the diurnal rectifier on remote marine boundary-layer stations. A basic requirement for such an analysis is that the CO₂ observations be traceable to the calibration scales maintained by the laboratories responsible for measurements from the network of marine boundary-layer stations. Summertime continental/marine concentration differences for various species would constrain the magnitude and extent of the diurnal rectifier, while observed seasonal variations in these differences would provide quantitative information on the influence of the seasonal rectifier on interhemispheric gradients measured at background stations.

Accurately distinguishing terrestrial and industrial CO₂ signals, and relating them to background-station values, will require high-accuracy measurements at fine spatial and temporal resolutions in and around the boundary layer. The required accuracies and precisions for CO₂ are on the order of a few tenths of a ppm (see Figure 2), while the requirements for CO are approximately 50 times as strict. Although such measurements are far from trivial on an aircraft platform, available instruments can measure CO₂ with an accuracy of 0.1 ppm at 0.5 Hz [B. Daube, personal communication] and CO with an accuracy of 1.5 ppb at 1 Hz [Gerbig et al., 1999]. Measurements of O₂ concentration would need to have a precision of 1-2 per meg, approximately equivalent to 0.2-0.4 ppm CO₂, to provide additional constraints. While airborne O₂ measurements at this level may be possible using an ultraviolet absorption technique [Stephens, 1999], presently they are limited to flask-sampling methods.

4. HARVARD FOREST DATA

To test the ability of CO data to distinguish CO₂ signals from fossil fuel emissions and terrestrial vegetation, we can use concentrations of CO₂ and CO, and vertical fluxes for CO₂ and momentum measured continuously for several years at Harvard Forest, Massachusetts (42.5°N, 72.2°W). Potosnak et al. [1999] have carried out such an analysis. Here we briefly present a subset of their results that are most relevant to measuring rectifiers. Figure 3 shows hourly CO₂ data for January and July 1996, with monthly fits to the form

$$[CO_2] = a_0 + a_1[CO] + a_2F_p + \sum_{j=0}^{7} a_{3j} \delta_{jt}$$  (1)

The term $a_0$ is the monthly-mean regional background concentration from 0000 to 0300 local time, and $F_p$ is a flux parameter that accounts for CO₂ gradients that develop within the PBL from local exchange with the underlying canopy: $F_p = \phi/u^*$ where $\phi$ is the CO₂ vertical flux measured by eddy correlation and $u^*$ is the friction velocity. This assumed scaling implies that at zero local CO₂ flux, or infinite local momentum flux, the tower CO₂ concentration would equal that of the regional boundary-layer. The variables $a_{3j}\delta_{jt}$ represent diurnal CO₂ variations in the regional boundary layer, resulting from diurnal variations in regional CO₂ flux and boundary-layer height. The data are binned into 3-hour intervals corresponding to $f = 0$ through 7 and $\delta_{jt}$ is set to 1 when $j = f$ and otherwise to 0. The time-of-day parameters $a_{3j}$ are derived for each interval using a generalized linear model [Venables and Ripler, 1994], thus allowing the model to fit an arbitrary shape to the monthly-mean diurnal cycle [Potosnak et al., 1999].

Table 1 shows the results of the statistical fits from Figures 3a and 3b. The model fits the hourly data very well in every month with $r^2$ values typically between 0.7 and 0.9. The coefficient multiplying the local flux parameter ($a_2$), and the range of the day factors (amp($a_{3j}$)), both reflect the dominant influence of biogeochemical exchange during summer and its relatively small influence during winter. The CO₂:CO slopes shown in Figures 3b and 3d are fairly reproducible, but are slightly lower in summer due to the oxidation of hydrocarbons co-located with CO₂ emissions.

We have removed the influence of combustion on the CO₂ variance at Harvard Forest by substituting into equation (1) the CO concentration at the 20th percentile for the month, which gives a good estimate for the zonal mean background [Goldstein et al., 1995]. We removed proximate effects of surface exchange by inserting zero for the local flux parameter. Figure 4a shows results given by the fitted formula for two different times of day with these parameters. These monthly values represent
the background concentrations in the PBL plus the influence of the monthly-mean regional diurnal cycle due to variations in biotic exchange and PBL height. We have compared these derived concentrations to CMDL data from Cold Bay, Alaska (55.2°N, 162.7°W), an upstream marine boundary-layer site of comparable latitude. As Figure 4a shows, this method of removing the effects of industrial and local canopy exchange using monthly statistical fits produces plausible background seasonal CO₂ cycles for a continental station [Potosnak et al., 1999].

Wintertime CO₂ in the PBL over Harvard Forest appears to be slightly higher than at Cold Bay at all times of day (Figure 4a), as expected due to net respiration. During summer when the terrestrial biota have more of an influence, early morning CO₂ values are higher and afternoon values are lower at Harvard Forest than at Cold Bay (Figure 4a). Figure 4b compares the observed background differences between Harvard Forest and Cold Bay to differences predicted by the TM2 model. Even though the biota are a strong net sink at this time, the 24-hour mean concentrations at 30 m in
Harvard Forest appear higher than at Cold Bay. The model predicts a depression of 7 ppm in 24-hour mean CO$_2$ at this height in midsummer, which is considerably different than the observations. This discrepancy is likely the result of a strong diurnal rectifier that is not reproduced in the TM2 model.

These analyses are encouraging for the use of CO data to account for industrial influences on airborne CO$_2$ data. However, there are still considerable uncertainties concerning the sources for CO at Harvard Forest [Potasnak et al., 1999], as well as the regional variability of industrial CO$_2$:CO ratios that would be encountered during continental scale measurements. The ability to derive continental background CO$_2$ concentrations from tower measurements would allow significant improvements to global inversion calculations. However, airborne measurements at different times of year over Harvard Forest would first be needed to investigate the spatial and temporal representativeness of the $a_0$ values calculated by Potasnak et al. [1999].

5. AMAZON DATA

Data obtained in the Amazon region from 1982-87 provide additional preliminary insights into rectifier effects, and information to guide future measurements. Figure 5 [Keller and Wofsy, unpublished data, 1983-84] shows flask measurements from November, 1983 to July, 1984 obtained in the interior of Amazonia near Manaus and at the same latitude (3°S) at the coast near Fortaleza. Samples were obtained in mid-morning or mid-afternoon in well-ventilated conditions. The data for CO$_2$ show higher concentrations in the interior (downwind), similar to the observations for CH$_4$ and N$_2$O, even though these gases are emitted in the region while CO$_2$ is being taken up at the time of sampling. This appears to be a manifestation of the diurnal rectifier, however as noted above, the possibility of an inland CO$_2$ source cannot be ruled out from surface data alone.

Figure 6 shows airborne observations of CO$_2$ over Amazonia made during the Atmospheric Boundary Layer Experiment 2B (ABLE2B). We have aggregated data from 15 flights near Manaus. The surface CO$_2$ excess in the morning was as high as 20 ppm [Wofsy et al., 1988], which was well simulated by the model of Denning et al. [1996]. As Figure 6 shows, the low values that characterize altitudes above 1 km were generated in the PBL in late afternoon, when the PBL height was maximum and CO$_2$ concentrations minimum (compare to Figure 1). These low concentrations are stranded aloft when PBL mixing ceases in the evening. The mean concentration was lower than Christmas Island, an equatorial marine boundary-layer site, at most altitudes, but CO$_2$ concentrations were higher than adjacent coastal regions below 2 km [Wofsy et al., 1988], consistent with a strong diurnal rectifier.

We can again test the ability to separate the influence of combustion from canopy exchange on CO$_2$ variability using a statistical model fit to the 9500 simultaneous CO and CO$_2$ measurements in this data set. We binned the data from all 15 flights by altitude ($\Delta z = 0.2$ km, range 0.2-6 km) and time ($\Delta t = 1$ hr, range 8-17 hr) to obtain data classified with a factor ($a_{ij}d_{ij}$) having 300 possible values. There were ample measurements to define the diurnal variation at altitudes below 3 km, but more sparse diurnal coverage above. The overall fit to the form

$$[\text{CO}_2] = a_0 + a_1[\text{CO}] + \sum_{j=0}^{29} a_{2j}d_{2j}$$

has residual standard error of 2 ppm and an $r^2$ of 0.6, with about half of the residual error due to sensor variance inherent to this early instrument. The accurate representation of the ABLE2B data by this linear model can again provide a basis for separating background ($a_0$), combustion-derived ($a_0 + a_1[\text{CO}$)], and biogenic (setting [CO] to background value) CO$_2$.

The fit to equation (2) provides an estimate for the CO$_2$ from combustion and other oxidation processes of $71 \pm 2$ mol CO$_2$:mol CO, similar to the values obtained from Harvard Forest data (Table 1). This is likely a result of variable mixing of Northern Hemisphere air at a time when fossil fuel burning dominates the interhemispheric gradient. Harris et al. [1990] inferred that much of the CO variance in ABLE2B was associated with episodic influx of polluted air from the Northern Hemisphere, and it is likely that the urban region of Manaus also contributed to the observed signal. The

<table>
<thead>
<tr>
<th>parameter</th>
<th>January 1996</th>
<th>July 1996</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_0$</td>
<td>354.9</td>
<td>358.7</td>
</tr>
<tr>
<td>$a_1$</td>
<td>0.083</td>
<td>0.059</td>
</tr>
<tr>
<td>$a_2$</td>
<td>0.005</td>
<td>0.030</td>
</tr>
<tr>
<td>$\text{amp}(a_{2j})$</td>
<td>0.85</td>
<td>19.45</td>
</tr>
<tr>
<td>$r^2$</td>
<td>0.95</td>
<td>0.74</td>
</tr>
</tbody>
</table>
observed CO$_2$ : CO ratio is notably higher than the value of 11.7 obtained from biomass burning plumes in the region during ABLE2A [Andreae et al., 1987], however ABLE2B took place during the wet season when there was very little biomass burning in the region. The other major source identified for CO during this period, oxidation of isoprene, produces about 1:1 CO$_2$ : CO and appears to have a negligible effect on the correlated variance of CO$_2$.

6. A SIMPLE BOUNDARY-LAYER MODEL

Intensive airborne measurements such as the ABLE2B data just shown can provide reliable estimates of regional CO$_2$ and CO fluxes. Wofsy et al. [1988] used a direct mass-balance approach for the CO$_2$ column amount in the lower atmosphere to compute a midday uptake of 9 ± 4 kgC/ha/hr, indistinguishable from measurements by eddy correlation obtained in a subsequent experiment [Fan et al., 1990]. Future attempts at measuring regional fluxes would benefit from measurements of additional PBL parameters such as heights, entrainment rates, and buoyancy fluxes, as well as from the implementation of an improved conceptual framework.

Following the work of Tennekes [1973], McNaughton and Spriggs [1986], Raupach et al. [1992], Raupach and Finnigan [1995], Denmead et al. [1996], and others, we express the variation of CO$_2$ in a well-mixed PBL over time by:

$$\frac{\partial C_{bl}}{\partial t} = \frac{\partial v}{\partial t} - \langle w \rangle (C_u - C_{bl}) + S \quad \text{for} \quad \frac{\partial v}{\partial t} - \langle w \rangle > 0$$

$$S \quad \text{for} \quad \frac{\partial v}{\partial t} - \langle w \rangle \leq 0$$

where $h$ is the boundary-layer height, $C_{bl}$ is the CO$_2$ concentration in the boundary layer, $t$ is time, $\langle w \rangle$ is the regional-mean vertical wind component, $C_u$ is the background CO$_2$ concentration, and $S$ is the surface exchange flux. This model is two-dimensionally Lagrangian in the sense that it tracks the horizontal motion of a column of air, but accounts for the effect of vertical advection ($\langle w \rangle$) across the concentration jump at the top of the boundary layer. In a typical continental high-pressure air mass, $\langle w \rangle$ represents the mean subsidence velocity and, as demonstrated by Raupach and Finnigan [1995], can be a significant term in boundary-
layer budgeting. The lower condition in Equation (3) represents times when either uplifting velocities exceed the rate of boundary-layer growth or boundary-layer collapse occurs faster than the rate of subsidence, during which the background CO$_2$ concentration can not influence that in the boundary layer.

Figure 7a shows variations in $h$ and $S$ over a 24 hour period, estimated from CO$_2$ (Figure 6), O$_3$, and H$_2$O measurements made during ABLE2B. The timing of both these parameters is linked to daily variations in solar intensity; CO$_2$ uptake is roughly proportional to the amount of photosynthetically active radiation and the PBL grows and shrinks with solar-driven buoyancy flux variations. It is this naturally tight correlation which drives the diurnal, and the seasonal, rectifiers.

Figure 7b shows the CO$_2$ concentration predicted at several heights by equation (3) using the values in Figure 7a as input. $C_{bi}$ increases throughout the night, then decreases rapidly with the onset of photosynthesis. As the PBL grows, air from increasing heights is entrained as indicated by the concentration jumps in Figure 7b. Even for a net balanced source, $C_{bi}$ only drops slightly below the background concentration for 6 hours in the afternoon. It is easy to see from Figure 7b how the 24-hour mean surface concentration could be much higher than background, and how difficult it could be to estimate net fluxes from surface data alone.

Airborne data provides the advantage of being able to monitor concentration variations in total or partial atmospheric columns. Figure 7c shows results from equation (3) for the CO$_2$ concentration, $C_{max}$, averaged from the surface to the height of daily maximum PBL growth. By comparison, $C_{bi}$ is averaged from the surface to the instantaneous PBL height. This column-averaged concentration is not affected by variations in PBL height, and reflects primarily the integral of the exchange rate, $S$:

$$h_{max} \frac{\partial C_{max}}{\partial t} = - \langle w \rangle (C_u - C_{max}) + S$$  \hspace{1cm} (4)

assuming $\langle w \rangle$ is negative and that the subsiding air spreads equally at all heights below $h_{max}$. While the significance of the subsidence term $\langle w \rangle$ to $C_{bi}$ in Equation (3) depends on its magnitude relative to the change in boundary-layer height $\langle \partial h/\partial t \rangle$, $\langle w \rangle$ is the only thing other than the surface flux $(S)$ that can modify $C_{max}$ in Equation (4). By integrating equation (4) over 24 hours we get

---

**Figure 5.** Concentrations of (a) CO$_2$, (b) N$_2$O, and (c) CH$_4$ observed in the Amazon. Measurements are shown from flasks collected during 1983-4 on the east coast of Brazil in Fortaleza (open circles) and 1000 km downwind at the same latitude (3°S), near Manaus (black squares) [Keller and Wofsy, unpublished data, 1983-84].
which demonstrates that straightforward estimates of the net exchange flux, \( \langle S \rangle \), may be made by measuring the average subsidence rate and the column abundance of \( \text{CO}_2 \). Furthermore, as Figure 7c shows, this doesn’t necessarily require intensive 24-hour monitoring; a reasonable estimate of \( \langle C_{\max} \rangle \) might be obtained by interpolating column averages made at sunrise and sunset. The application of this 1-D model is subject to uncertainties regarding the assumption of consistent subsidence into and lateral spreading within the boundary layer. While extensive airborne \( \text{CO}_2 \) measurements would benefit from this conceptual framework, at the same time they would provide important data for validating various flux-estimate techniques [Denmead et al., 1996].

\[
\langle C_{\max} \rangle - \langle C_o \rangle = \frac{\langle S \rangle}{\langle w \rangle}
\]  

7. THE \( \text{CO}_2 \) BUDGET AND RECTIFICATION AIRBORNE STUDY

We plan to implement the measurement strategies discussed above during the \( \text{CO}_2 \) Budget and Rectification Airborne Study (COBRA). Our approach will be to obtain extensive measurements of the vertical and horizontal distributions of \( \text{CO}_2 \), \( \text{CO} \), \( \text{O}_2 \), \( \text{O}_3 \), \( ^{13}\text{CO}_2 / ^{12}\text{CO}_2 \) and other tracers in and above the PBL across North America and the adjacent ocean regions during two month-long field campaigns; one during the summer of 2000, and the other during the winter of 2000-2001. Preliminary data collected on test flights during the summer of 1999 will help to plan the later measurements. The platform will be the University of North Dakota Citation II aircraft. We plan continuous measurements of \( \text{CO}_2 \) using an infra-red analyzer [Boering et al., 1994], of \( \text{CO} \) using a resonance fluorescence instrument [Gerbig et al., 1999], and of \( \text{O}_3 \) using a UV
instrument [L. Avallone, personal communication] with approximate vertical and horizontal resolutions of 5 and 150 meters, respectively. We will take flask samples for the measurement of CO$_2$ and O$_2$/N$_2$ ratios at Scripps and for the measurement of CO$_2$, CH$_4$, CO, H$_2$, SF$_6$, N$_2$O, $^{13}$CO$_2$/$^{12}$CO$_2$, and $^{18}$O/$^{16}$O/C$^{16}$O$_2$ at CMDL.

As for the Harvard Forest and ABLE2B analyses above, we plan to develop statistical models of our data that enable the use of tracer-tracer correlations to separate the influence of terrestrial biotic exchange, fossil fuel combustion, and oceanic exchange on the observed CO$_2$ variations. The resulting summertime continental-scale terrestrial CO$_2$ signals will quantitatively measure the diurnal rectifier, while comparisons between our summer and winter measurements will constrain the seasonal rectifier. We plan to use output from the Denning et al. [1996] model and other coupled terrestrial-atmospheric models to help plan our flight patterns and interpret the resulting data.

In addition to spatially extensive continental-survey flights, we also plan temporally intensive diurnal observations in and above the planetary boundary layer in selected regions. These flights will extend over 100-300 km, centered above the WLEF tower in Wisconsin [Bakwin et al., 1998] and Harvard Forest [Goulden et al., 1996], where continuous measurements of flux and concentrations of CO$_2$ and CO are currently made. At the WLEF tower we will have measurements of all the terms in equations (3)-(5), providing an ideal framework to elucidate the functioning of the diurnal rectifier, to investigate the feasibility of determining regional fluxes using aircraft observations, and to define the optimal suite of tracer and ancillary data.

Together, the continental-survey and diurnal studies will characterize the spatial and temporal variability of CO$_2$ concentrations over North America. This information will aid in assessing the design and feasibility of an airborne, flask-based, carbon cycle observing network such as Carbon America [Tans et al., 1996]. It will also allow us to investigate other protocols for calculating regional to continental scale CO$_2$ fluxes. For example, by vertically integrating the observed terrestrial and fos-
sil fuel CO₂ components, we can define ratios between these influences for specified regions across North America. By multiplying these terrestrial-industrial ratios by the relatively well known fossil fuel source, we can estimate net terrestrial CO₂ fluxes for those regions.

Our planned suite of measurements will have additional applications beyond CO₂ budget and rectification questions. The O₂/N₂ data, with CO₂, will provide the first direct measurement of photosynthetic and respiratory quotients over large regions. These ratios, assumed equal to 1.1 based on laboratory incubation studies [Severinghaus, 1995], are significant for ecosystem studies and critical to the inversion of CO₂ and O₂ data to obtain regional and global carbon budgets [Keeling et al., 1996]. The observed ¹³CO₂/¹²CO₂ and C₁₈O₁₆O/C₁₆O₂ gradients will provide new measurements of the isotopic signatures of terrestrial CO₂ exchange on continental scales. The CH₄, N₂O, and O₃ measurements will help to improve our current understanding of the global budgets for these species.

8. CONCLUSION

Inverse calculations of surface CO₂ fluxes are currently subject to significant uncertainties associated with the representation of seasonal and diurnal rectifier effects. The data from Harvard Forest and the Amazon presented here suggest that even during times of net CO₂ uptake, the diurnal rectifier maintains continental surface concentrations higher than those at marine boundary-layer sites. However, additional measurements are clearly needed to quantify vertical and horizontal CO₂ gradients resulting from both the diurnal and seasonal rectifier effects. We have shown that airborne measurements of CO₂, CO, O₂, and other tracers might allow the independent identification of continental-scale terrestrial CO₂ signals. We have also presented a simple boundary-layer model that may provide a framework for using airborne measurements to calculate regional CO₂ fluxes, and to investigate rectification processes.

Airborne measurements have the potential to bridge the gap between tower-based flux measurements and global scale carbon budget constraints, and to reduce large uncertainties in the relationships between surface CO₂ concentrations, rates of mixing and advection, and surface CO₂ fluxes, which currently limit our ability to define the present carbon cycle. Such measurements can also contribute to designing optimal strategies for measuring regional and continental scale fluxes. We have outlined plans for the CO₂ Budget and Rectification Airborne Study, which incorporates these goals.

An important component of this and other studies will be comparison of airborne observations to model predictions: 1) to test parameterizations of vertical mixing and biotic fluxes, 2) to help define continental net sources and sinks, and 3) to assess the impact of rectification gradients on inverse calculations. Once coupled terrestrial-atmosphere models are shown to adequately represent rectification effects, we can have much greater confidence in estimates of surface CO₂ fluxes from inverse calculations.

NOTATION

\[ a_0 \] Monthly-mean regional background CO₂ concentration in Equations (1) and (2)
\[ a_1 \] Coefficient multiplying CO concentration in Equations (1) and (2)
\[ \phi \] Vertical CO₂ flux
\[ u^* \] Friction velocity
\[ F_p \] Flux parameter equal to \( \phi/u^* \) in Equation (1)
\[ a_2 \] Coefficient multiplying flux parameter in Equation (1)
\[ \delta_{ij} \] The Kronnecker delta in Equations (1) and (2)
\[ a_{3j} \] Coefficient multiplying the Kronnecker delta in Equation (1)
\[ a_{2j} \] Coefficient multiplying the Kronnecker delta in Equation (2)
\[ h \] Height of the boundary layer
\[ t \] Time
\[ C_{bl} \] Average CO₂ concentration within the boundary-layer
\[ C_u \] Background CO₂ concentration
\[ C_{(w)} \] Regional-mean vertical wind component
\[ S \] Surface CO₂ flux
\[ h_{max} \] Daily maximum height of the boundary-layer
\[ C_{max} \] Average CO₂ concentration below \( h_{max} \)

Acknowledgments. B. Stephens was supported by an NSF Graduate Research Fellowship during the preparation of this manuscript. The COBRA project has been jointly funded by DOE, NOAA, NASA, and NSF.

REFERENCES


Dargaville, R. J., and Simmonds, I., Calculating CO2 fluxes by data assimilation coupled to a three dimensional mass balance inversion, this volume, 1999, American Geophysical Union.


Taguchi, S., Synthesis inversion of atmospheric CO2 using the NIRE chemical transport model, this volume, 1999, American Geophysical Union.


Britton B. Stephens, Marine Research Division, Scripps Institution of Oceanography, La Jolla, California, USA. (email: britt@ucsd.edu)

Steven C. Wofsy, Department of Earth and Planetary Science, Harvard University, Cambridge, Massachusetts, USA.

Ralph F. Keeling, Marine Research Division, Scripps Institution of Oceanography, La Jolla, California, USA.

Pieter P. Tans, Climate Monitoring and Diagnostics Laboratory, NOAA, Boulder, Colorado, USA.

Mark J. Potosnak, Lamont-Doherty Earth Observatory, Columbia University, Palisades, New York, USA.