Measurements of carbon dioxide on very tall towers: results of the NOAA/CMDL program

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ABSTRACT

Measurements of carbon dioxide (CO_2) mixing ratios have been carried out since 1992 on a 610-m tall communications tower in North Carolina and since 1994 on a 447-m tall tower in Wisconsin. The data provide insights into the influence of pollution (fossil fuel combustion), biological exchange, boundary layer dynamics, and advective transport on CO_2 mixing ratios over the continents. In this paper, we provide an overview of the data, describe access to the data, and suggest ways in which these results could be used to improve simulations of the global carbon cycle. In particular, the data will be very useful to constrain model estimates of covariance between terrestrial ecosystem fluxes of CO_2 and diurnal and seasonal variations of planetary boundary layer mixing.

1. Introduction

Long-term observations of carbon dioxide (CO_2) mixing ratios in the atmosphere have provided a basis for improved understanding of the global carbon cycle. Observed spatial gradients of CO_2 have been used to calculate net surface exchange on very large spatial scales (e.g., in large latitude bands or on the scale of continents and ocean basins) by means of inverse modeling (Tans et al., 1990; Enting and Mansbridge, 1987; Denning, 1994; Fan et al., 1998). These models rely on simulated atmospheric transport to translate observed mixing ratio distributions into spatially resolved surface fluxes. Since emissions of CO_2 from combustion of fossil fuel are relatively well quantified on a rather fine spatial scale ($1° \times 1°$, Andres et al., 1998) it is possible to compute the fluxes resulting from exchange between the atmosphere and the terrestrial bio-

sphere and oceans. Several recent studies have concluded that a large net sink for CO_2 must exist at temperate latitudes of the Northern Hemisphere to balance the global carbon cycle and that a significant fraction of this must be due to net uptake by the terrestrial biosphere (Tans et al., 1990; Denning, 1994; Ciais et al., 1995; Fan et al., 1998). However, the paucity of observations in the regions where net uptake appears largest, the northern temperate continents, places a severe restriction on the ability of model studies to pinpoint the temporal and spatial character of the sink. Quantitative understanding of the carbon cycle on continental or finer spatial scales, and annual time scales, is an important first step towards prediction of future atmospheric CO_2 levels under a given scenario of fossil fuel combustion and/or land use change. In addition, the 1992 United Nations Framework Convention on Climate Change (text of the Convention is available on the web at http://www.unfccc.de/index.html) requires signatory nations to inventory sources and

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sinks for greenhouse gases within their borders, which requires a regional (as opposed to global) approach.

The net exchange of CO_2 between the atmosphere and a terrestrial ecosystem can be directly measured and monitored over long periods (years) by direct flux methods such as eddy covariance (Wofsy et al., 1993; Grace et al., 1995; Goulden et al., 1996). These studies are limited in their spatial scope addressing scales of only a few hectares, but are very useful for relating fluxes to biophysical factors including climate, soil properties, and vegetation dynamics that control carbon exchange with the biosphere. Other approaches are necessary to quantify CO_2 exchange on larger regional scales and to verify extrapolations from local flux measurements (Tans et al., 1996b).

Several monitoring studies have been initiated in recent years to observe the climatology of CO_2 mixing ratios over continental regions. Levin (1987) analyzed CO_2 and radon-222 data from a mountain top site in southern Germany to estimate regional terrestrial exchange. The study was extended by Levin et al. (1995) and Schmidt et al. (1996). Kuc (1991) reported on continuous measurements since 1983 of CO_2 and the ^{14}C and ^{13}C composition of CO_2 in 2-week-integrated samples obtained near Krakow, Poland. Haszpra (1995) reported continuous observations of CO_2 from a rural site in Hungary.

Our laboratory has taken steps to extend the coverage of our global network for monitoring CO_2, CH_4, and CO to include greater representation of continental areas. We have recently added several flask sampling stations in areas near to or on the continents, but where local sources and sinks for these species are minimal, such as the Gobi Desert of Mongolia, the Great Basin desert of the United States, a ferry boat on the Baltic Sea, Malta (Mediterranian Sea), the Tae-Ahn Peninsula on the west coast of South Korea, and others (Tans et al., 1996a). We initiated a program to determine vertical profiles of CO_2, CH_4, and CO mixing ratios through the continental lower troposphere by flask sampling aboard small aircraft and since 1992 have made weekly to bi-weekly measurements over northcentral Colorado (Tans et al., 1996a). Also, we have begun continuous in situ monitoring of CO_2 and other tracers on existing tall communications towers in the United States. By using very tall towers we are able to measure CO_2 mixing ratios representative of the continental planetary boundary layer. The experimental design and first results from our observations of CO_2 on a 610-m tall tower in North Carolina (NC) were reported by Bakwin et al. (1995). In this paper we give further results from continuing observations at the NC tower and from a 447-m tall tower in Wisconsin (WI).

Data from the North Carolina and Wisconsin towers presented in this paper are available via anonymous ftp. The on-line data products are described in Subsection 3.5.

2. Experimental

2.1. Study sites

Data were obtained from two television and radio transmitter towers: a 610-m tall tower located in a rural area of eastern North Carolina (35.37°N, 77.39°W, 9 m above sea level) and a 447-m tall tower located in a forested area of northern Wisconsin (45.95°N, 90.27°W, 472 m above sea level). Both towers are guyed steel scaffolding with faces 3–4 m wide. The region surrounding the NC tower has been described in detail elsewhere (Bakwin et al., 1995; Hurst et al., 1997).

The WI tower is located in the Chequamegon National Forest, 14 km east of Park Falls, Wisconsin (population 3200). The region is in a heavily forested zone of low relief. The Chequamegon National Forest covers an area approximately 325,000 ha, and the dominant forest types are mixed northern hardwoods (85,000 ha), aspen (75,000 ha), and lowlands and wetlands (60,000 ha). The region immediately surrounding the tower is dominated by boreal lowland and wetland forests typical of the region. Much of the area was logged, mainly for pine, during 1860–1920 and has since regrown (J. Isebrands, U.S. Department of Agriculture Forest Service, personal communication, 1995). The region for >50 km around the WI tower is dominated by forest with some small farms and villages. At greater distances, especially to the south, the land is largely cleared for agriculture.

The regions around the two towers differ greatly in human population density. The population density of Pitt County, which contains the NC tower, averaged 64 km^{-2} in 1990, and the popula-

Table 1. *Summary of climate for stations near the North Carolina (NC) and Wisconsin (WI) towers*

	NC	WI
annual P (cm)	130.2	79.5
annual average T (°C)	16.4	4.1
January average T (°C)	5.5	−12.9
July average T (°C)	25.9	18.9

P = precipitation; T = temperature. Data from Vose et al. (1992) for Kinston, NC (20 km southwest of the NC tower), and Minocqua, WI (40 km east of the WI tower). Averages for 1950–1987.

tion density of Price County, which contains the WI tower, averaged about 5 km^{-2} (data from the U.S. Census Bureau web site, 1997). These population densities are roughly typical of the regions within 50 km or more of the towers.

The tower sites are also quite different in their climates (Table 1). The WI tower is located in a zone of cold, temperate, midcontinental climate, while the region around the NC tower experiences a climate that is moderated by its proximity to the Atlantic Ocean 80 km to the southeast. The amplitude of the seasonal cycle of monthly mean temperature is much larger at the WI tower than at the NC tower, while annual precipitation is ≈65% greater at the NC tower.

2.2. Measurements

Measurements of CO_2 mixing ratios began in June 1992 at the NC tower and in October 1994 at the WI tower. Observations at the NC tower are described in detail by Bakwin et al. (1995). Briefly, at the NC tower CO_2 is measured continuously at 51, 123, and 496 m above the ground by non-dispersive infrared spectroscopy (LiCor, Lincoln, Nebraska, model 6251). The reference cells of the analyzers are flushed with a compressed gas standard containing ≈330 ppm (parts per 10^6 by mole ratio) CO_2 in air. The sample air is dried to a dewpoint of −25° by passing the air first through a refrigerated (≈5°C), continuously purged liquid water trap, then through a Nafion drier (Permapure, Toms River, New Jersey, model MD-250–72P). A multiport valve is used to control calibration and sampling sequences. A calibration gas containing about 330 ppm CO_2 is introduced to each LiCor every 36 min in order

to track short-term drifts in the instrument background that are mainly caused by variations in room temperature. About every three hours we calibrate with a sequence of four standards containing approximately 330, 360, 390, and 420 ppm CO_2. The calibration gases are prepared in our laboratory as described by Kitzis (1998) and are referenced to the WMO mole fraction scale (Zhao et al., 1997a). Precision of the calibrations is estimated to be 0.1 ppm, and accuracy is about 0.2 ppm (Zhao et al., 1997a).

Procedures for CO_2 measurements at the WI tower are very similar to those at the NC tower. The measurement heights at WI are 11, 30, 76, 122, 244, and 396 m above the ground. We have 2 CO_2 analyzers for mixing ratio measurements: one analyzer measures air from 396 m continuously while the other analyzer cycles through all 6 measurement levels. 2 min are required to obtain a steady reading after each valve switch, so a full profile is carried out in 12 min.

Our PC-based data acquisition and control system has been described in detail by Zhao et al. (1997b) and Zhao and Bakwin (1997). Raw data are transferred automatically every day from each tower site to our laboratory via modem.

We measure a suite of other trace gas species at both towers by using in situ gas chromatography (GC). The species measured are CO (carbon monoxide), H_2 (hydrogen), CH_4 (methane), CCl_2F_2 (chlorofluorocarbon 12, CFC-12), CCl_3F (CFC-11), CCl_2FCClF_2 (CFC-113), $CHCl_3$ (chloroform), CH_3CCl_3 (methyl chloroform), CCl_4 (carbon tetrachloride), C_2Cl_4 (tetrachloroethene), N_2O (nitrous oxide) and SF_6 (sulfur hexafluoride). Hurst et al. (1997) discuss in detail the methodology used for the GC measurements and initial results for the NC tower, and Bakwin et al. (1997) analyzed the GC data from the NC tower to extract pollution signatures for these species. The GC observations are of particular value for identifying pollution sources of CO_2, since the CFCs, other halocarbons, and SF_6 are only emitted from anthropogenic sources and their mixing ratios are highly correlated in pollution plumes. Further, CO is a key tracer for combustion.

Once per week, a pair of flask samples is collected from the highest level on each tower. The samples are analyzed at our laboratory in Boulder for mixing ratios of CO_2, CH_4, H_2, CO, N_2O and SF_6 (analyses for N_2O and SF_6 were

started in 1996) and by the Stable Isotope Laboratory of the Institute for Arctic and Alpine Research at the University of Colorado for the $^{13}C/^{12}C$ and $^{18}O/^{16}O$ isotopic ratios of CO_2 (Trolier et al., 1996). The $^{13}C/^{12}C$ data from the towers and other sites in our flask sampling network have been analyzed by Bakwin et al. (1998) to determine the isotopic signature of exchange between the atmosphere and the terrestrial biota.

In addition to the trace gases, we measure meteorological parameters on the towers including wind speed and direction, temperature, and humidity at 51, 123, and 429 m on the NC tower (note: in Bakwin et al., 1995, the top level for meteorological observations is incorrectly given as 404 m), and at 30, 122, and 396 m on the WI tower. On the WI tower three-dimensional wind data are collected using sonic anemometers, and the data are used to calculate surface fluxes of CO_2, H_2O, sensible heat, and momentum by the eddy covariance method (Davis et al., 1996). Analysis of the flux observations will be given in future publications. At both towers we also measure photosynthetically active photon flux density (LiCor model 190S quantum sensor) and rainfall using a tipping bucket rain gauge.

3. Results and discussion

Initial results from observations at the NC tower are given by Bakwin et al. (1995). In those results CO_2 mixing ratios show strong diurnal and seasonal cycles, as well as variability on a synoptic scale. These features of the time series from both towers are discussed further in the following sections.

3.1. Diurnal cycles

Fig. 1 summarizes the diurnal cycles at the NC and WI towers for January and July. In order to remove the influence of synoptic variability we have subtracted for each day the 24-h mean mixing ratio for the top tower level (496 m at NC, 396 m at WI) from observations at all levels for that day.

The diurnal cycle of CO_2 near the ground is driven by the diurnally varying biological source/sink and by dynamics of the planetary boundary layer (PBL). During the daytime in summer, photosynthetic uptake of CO_2 dominates over respiration, and the PBL is well mixed by convection and wind shear to typically 1–2 km depth. At our tower sites CO_2 mixing ratios in the mid-PBL (400–500 m) during the afternoon in summer are 2–3 ppm lower than the 24-h mean. We also observe a 1–3 ppm decrease in CO_2 nearer the ground, reflecting uptake by the vegetation. Bakwin et al. (1995) showed that CO_2 mixing ratios at 496 m on the NC tower in the afternoon are likely to be within a few tenths of a ppm of the column mean for the entire PBL. A similar result can be shown for the 396 m level on the WI tower. At night, the PBL is stably stratified and shallow (typically < 150 m), while respiration and pollution provide sources for CO_2. Mixing ratios of CO_2 build up to very high levels beneath the nocturnal temperature inversion. Our data indicate that, on average, some exchange occurs through the inversion and there is a gradual increase in CO_2 at levels > 200 m during the night, though on many nights this does not occur and mixing ratios above the inversion remain fairly constant. In summer the diurnal cycle at the highest sampling levels is dominated by a transient increase of CO_2 during mid-morning as the PBL grows through those levels. In winter the surface biological source/sink is greatly reduced, and daytime convective mixing is also suppressed. Vertical gradients are small throughout the day, and we observe little diurnal variation at 400–500 m above the ground (Fig. 1).

The influences of biological exchange and PBL dynamics on the diurnal cycle of CO_2 near the ground over terrestrial systems have been understood at least since the early 1950s from work by B. Huber (see review by Lieth, 1963). The covariance between PBL mixing depth and biological uptake (daytime) and release (night) of CO_2 results in higher daily mean mixing ratios near the ground than aloft (Denning et al., 1996b), as is clear from Fig. 1. By using existing transmitter towers we are able to extend continuous observations to much higher altitudes than previous studies. In particular, we are able to directly observe CO_2 mixing ratios in the middle of the convective (daytime) PBL where vertical gradients are relatively small (Bakwin et al., 1995) and well above the nocturnal near-surface inversion. Our observations should be particularly valuable to constrain and test parameterizations of PBL mixing in regional and
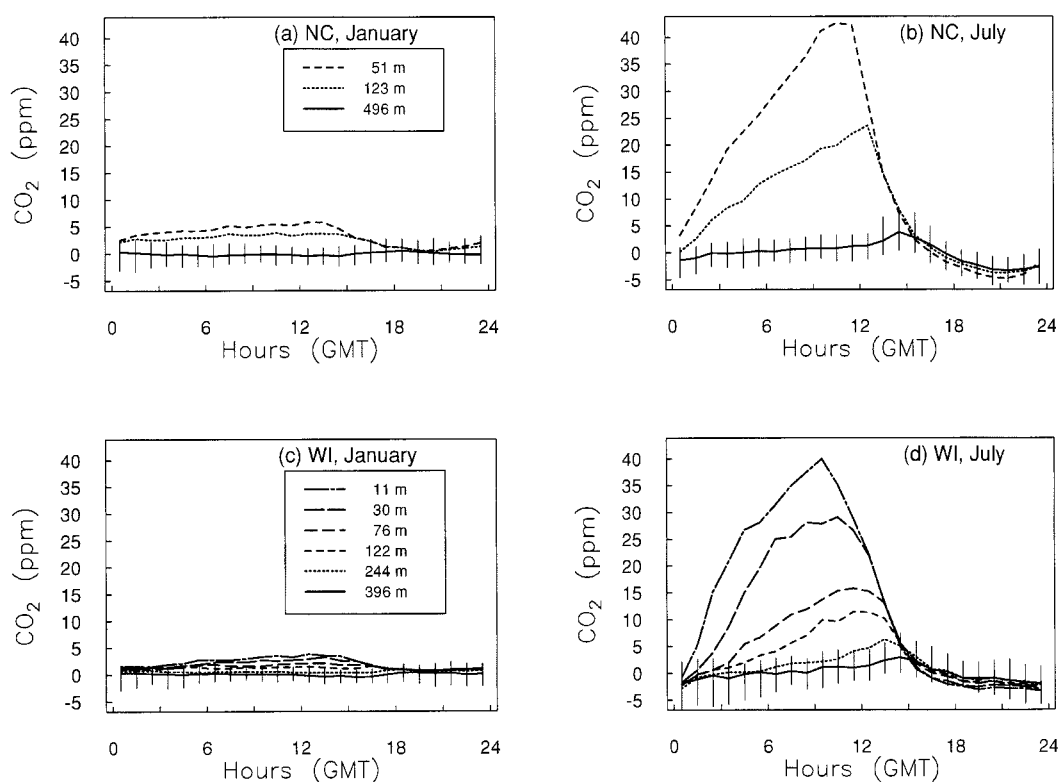
Fig. 1. Diurnal cycles of CO_2 (medians by hour) for each measurement level on the NC and WI towers for January and July. For each day the 24-h mean mixing ratio for the top tower level (496 m at NC, 396 m at WI) was subtracted from all observations for that day. Vertical lines show the range of the 16th and 84th percentiles for the top levels. Data for NC are composite for 1993–1997 and for WI are composite for 1995–1997. GMT is 5 and 6 hours ahead of standard time for NC and WI, respectively.

global models in which vertical resolution near the ground is typically rather course.

During the summer of 1995, observations of PBL depth and wind profile were made using a radar remote sensing system located about 8 km from the WI tower (W. Angevine, University of Colorado, unpublished data, 1995; see also Angevine et al., 1998). In July the convective PBL typically grew to about 1350 m depth by 1800 GMT (1200 local standard time) and the PBL depth was relatively constant during the afternoon. As shown in Fig. 1, mid-PBL (396 m) CO_2 mixing ratios also changed little, on average, during 1800–2200 GMT indicating that surface uptake of CO_2 by the forest was approximately balanced by entrainment of CO_2 into the PBL from aloft. Late in the afternoon CO_2 mixing ratios decreased somewhat (0.5–1.0 ppm), suggesting that PBL

mixing may have been suppressed at lower sun angles, while photosynthesis continued. At the NC tower CO_2 mixing ratios were also nearly constant, on average, for 3–4 h during the afternoon in summer, implying a similar balance between surface uptake and PBL entrainment. However, we have no observations of PBL depth near the NC tower. Analysis of the PBL development and CO_2 fluxes at the WI tower will be given in a future publication.

A comparison between the diurnal cycle of CO_2 observed at the NC tower and computed using the Colorado State University (CSU) global circulation model (GCM) coupled with the SiB2 biosphere model was presented by Denning et al. (1996b). In the model the PBL is a single well-mixed layer, and the PBL depth is calculated at each 6-min time step. The match between model

diurnal cycle in the PBL and our data from 51 m above the ground was best while the match with our data from 496 m was relatively poor. The reason for this is that the 51-m level is usually within, but the 496-m level is typically above, the nocturnal PBL. At 496 m at night CO_2 mixing ratios are generally much lower than the PBL mean, but during the daytime convective mixing is vigorous and mixing ratios at 51 m and 496 m are very similar; the afternoon vertical gradient (496 m–51 m) in summer is typically about 2 ppm (Bakwin et al., 1995), while the diurnal cycle amplitude at 51 m is ≈40 ppm (Fig. 1). Hence, CO_2 mixing ratios at 51 m are more representative of the 24-h column mean for the PBL. Good agreement for model calculated PBL CO_2 mixing ratios with our data from 76 m on the WI tower has also been shown by Denning et al. (1996c).

The daytime convective PBL is typically capped by a temperature inversion that often persists through the following night, sustaining a "relic" mixed layer in which turbulence is suppressed (Stull (1988); see also Wofsy et al. (1988)). Vertical exchange through the capping inversion is slow relative to mixing below that level, so the air below the inversion should tend to reflect the integrated influence of surface exchange. The residence time for air within the PBL is probably on the order of the timescale for synoptic disturbance (a few days). Our mixing ratio data from the highest tower levels are reasonably representative of mean values for the daytime PBL (Bakwin et al., 1995) and are not greatly affected by the large nighttime build-up below the shallow (≈100 m) nocturnal inversion. Therefore, to estimate the influence of surface fluxes on daily integrated CO_2 mixing ratios near the surface over the continents it may be appropriate to compare "background" mixing ratios with our data from 496 m (NC) and 396 m (WI) on the towers. Comparison of the seasonal cycles and annual means are presented in Subsections 3.2 and 3.3, respectively.

3.2. Seasonal cycles and short-term variability

Fig. 2 shows 24-h mean CO_2 mixing ratios at the 496-m level on the NC tower and the 396 m level on the WI tower, along with smooth curve fits to the data (Thoning et al., 1989), and reference curves that represent the marine boundary layer (MBL) average at the latitudes of the towers obtained from GLOBALVIEW — CO_2 (1997) (Masarie and Tans, 1995). In general the seasonal cycles at the towers lead those of the MBL as expected since the seasonal cycles at northern midlatitudes are driven primarily by terrestrial biology (Fung et al., 1983).

The seasonal cycles at the towers are much more variable than in the MBL. For example, the increase of CO_2 mixing ratios at the NC tower during the fall of 1993 was more gradual than in other years, leading to a somewhat wider summer minimum than usual, whereas the summer draw-down was deeper than usual (by about 2–3 ppm) in 1997. Also, the amplitude of the summertime drawdown at the WI tower was 4–5 ppm deeper in 1996 and 1997 than in 1995. These year-to-year differences may reflect changes in biological activity in the regions surrounding the towers or differences in transport of air to the measurement sites. Since climate anomalies are generally coherent over regional scales (thousands of km^2) it is reasonable to expect that such anomalies would result in year-to-year differences in seasonal patterns of CO_2 at the towers. For example, the summer of 1993 was extraordinarily dry and somewhat warmer than normal in the southeast United States (data from National Climatic Data Center, 1994). Possibly, these conditions led to lower-than-normal primary production for the region, followed by reduced respiration in fall, and hence a more gradual increase of CO_2. Also, the summer of 1995 was unusually warm in the eastern United States, particularly in the Great Lakes region, though precipitation was roughly normal in the region. The shallow summer minimum at the WI tower in 1995 could have been a result of lower biological productivity, greater respiration, or changes in transport associated with the anomalously warm weather in the region. Our ongoing flux measurements at the WI tower, coupled with observations of biophysical driving factors for changes in photosynthesis and respiration, will aide understanding of the processes that drive interannual changes in CO_2 mixing ratios observed at the site.

The average seasonal cycle amplitude for daily mean mixing ratios at 496 m on the NC tower for 1993–1997 is 16.9 ppm, and the average amplitude at 396 m on the WI tower for 1995–1997 is 22.4 ppm. The seasonal cycle amplitudes at the
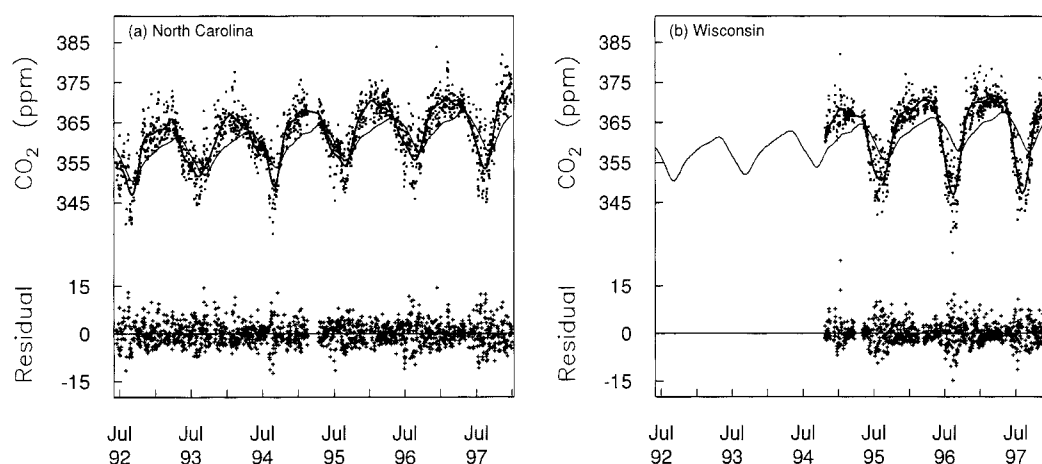
Fig. 2. Daily mean CO_2 mixing ratio at (a) 496 m on the NC tower, and (b) 396 m on the WI tower (upper panels). The data are fit with a smooth curve (heavy) consisting of a second order polynomial and four seasonal harmonics, and the residuals are smoothed with short (80 day) and long (667 day) cut-off filters (Thoning et al., 1989). The MBL reference curves (GLOBALVIEW — CO_2, 1997) for each latitude are also shown (thin line). The daily residuals from the smooth curve are shown in the lower panels.

towers are 75–100% larger than at MBL sites at the same latitudes (9.7 and 10.9 ppm, respectively).

The lower panels of Fig. 2 show a marked seasonal cycle of the short-term variability of daily mean CO_2 about the smooth curve fits. This is seen more easily in Fig. 3. The residual standard deviation (RSD) from the smooth curve fits peaks in midsummer at around 5 ppm and is much less in winter and spring, around 1–3 ppm, reflecting larger photosynthetic and respiratory fluxes in summer than in winter. In winter correlation with CO explains >80% of the variance in CO_2 at

Fig. 3. Standard deviation of residuals from smooth curve fits to daily mean CO_2 mixing ratios at the towers composited by month for all years of data, and for flask samples from Bermuda for 1993–1997.

these locations indicating that regional combustion is the main source of variability for CO_2, but in summer the correlation between CO and CO_2 is poor (Bakwin et al., 1994; Bakwin et al., 1995). Background (MBL) sites at northern midlatitudes, such as Bermuda (Fig. 3), show much less seasonality in the variability of CO_2 and the monthly RSD is typically around 1–2 ppm.

These observations have important implications for the "detectability" of net CO_2 exchange using the tower data. The short-term variability shown in Fig. 3 is a measure of the "noise" above which a signal from a regionally significant net surface flux would have to be detected (Tans et al., 1996b). For example, assuming that each 24-h average is an independent measure of continental values for CO_2 we can determine the summertime monthly mean CO_2 mixing ratio to within about 0.9 ppm (5 ppm RSD/$\sqrt{30}$ days), and we can determine wintertime monthly means to within about 0.4 ppm. Alternative methods would take account of covariance between CO_2 mixing ratio and other variables such as other tracers, wind trajectory, or PBL depth in order to improve the signal-to-noise ratio for quantifying a surface flux. We have conditionally averaged CO_2 mixing ratios and diurnal cycles within wind direction sectors (not shown) but this has not successfully reduced unex-
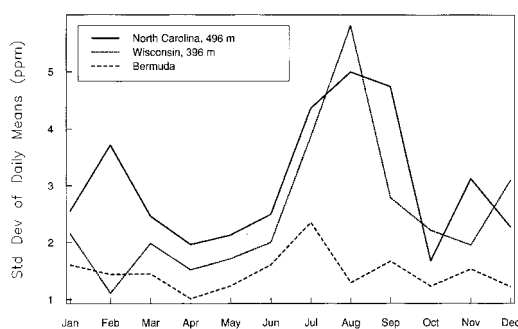
plained variance for summer. Trajectory calculations are beyond the scope of this study.

Smooth curve fits to daily mean CO_2 mixing ratios at each measurement level on the two towers are shown in Fig. 4. Seasonal cycles reflecting photosynthetic draw-down of CO_2 in summer and an increase due to net respiration and pollution in fall and winter are evident at the higher altitudes but are strongly damped at the lower levels (Fig. 5). In summer the build-up of CO_2 near the ground at night compensates for the large-scale drawdown so that the seasonal minimum is attenuated near the ground. With the exception of the 11 m level at WI the vertical gradient of seasonal cycle amplitude at both towers is roughly constant between years (Fig. 5), indicating that interannual differences in the seasonal cycle amplitude are driven by large-scale phenomena which affect all tower measurement levels equally, rather than by local changes in the vigor of vertical exchange or surface fluxes. These observations have implications for comparison of the data with models since models typically parameterize the PBL as a well-mixed layer of variable depth (shallow at night, deep during the daytime), while measurements are typically made at fixed altitudes which may be within or above the PBL depending on time of day and meteorological conditions.

The observed seasonal cycle amplitudes can be compared with predictions of global models: Fung et al. (1987) predicted seasonal amplitudes of about 10 and 15 ppm in the lowest model layer in the regions of the NC and WI towers, respectively, while Denning et al. (1996b) predicted PBL mean amplitudes of about 13 and 21 ppm for these locations. Comparison with model seasonal cycles should also take into account the phase of the seasonal oscillation. For example, in the model of Denning et al. (1996b) the maximum and minimum CO_2 mixing ratio in the PBL at the grid point containing our NC tower and resulting from annually balanced biospheric exchange (annual net flux = 0 at each grid point) occur in August and October, respectively (A. S. Denning, personal communication, 1997), while the observed seasonal maximum and minimum at 496 m are typically in January-March and August, respectively. The timing of the observed seasonal maximum and minimum at 51 m are more difficult to establish with confidence but appear to be similar to 496 m (Fig. 4). The seasonal cycle phase in the model results from the influence of the model climate on biospheric exchange, and the model simulates poorly the climate of this region (Denning et al., 1996a). The results of Denning et al. (1996b) for the region surrounding the WI tower are in much better accord with our data; in both cases the minimum CO_2 occurs in August and the maximum in late winter. This comparison illustrates the difficulty of interpretation of model
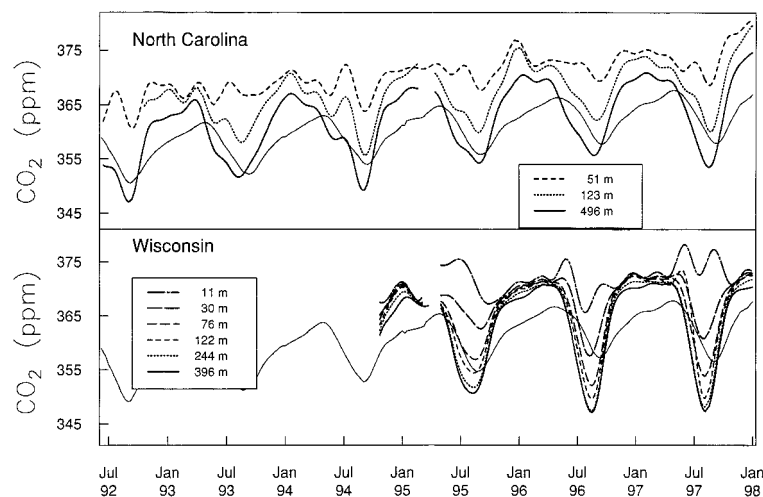

Fig. 4. Smooth curve fits (Fig. 2) to daily mean CO_2 mixing ratios at each measurement level on the NC and WI towers, and the MBL reference curves (thin line).
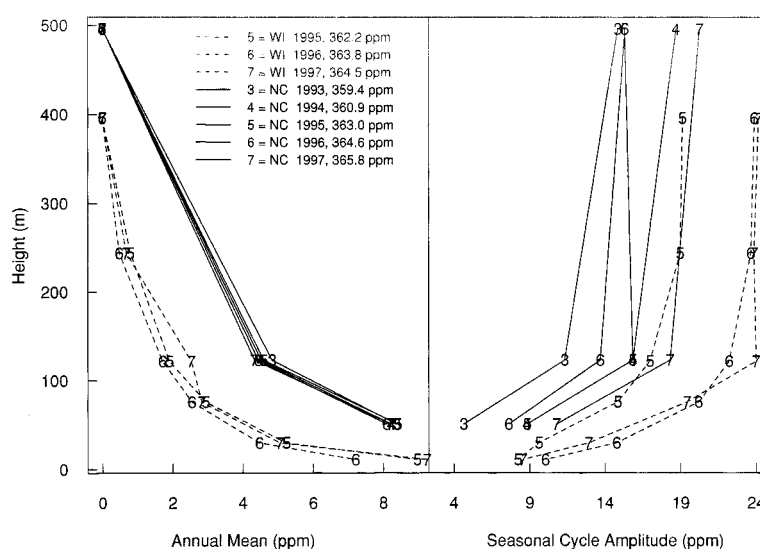
Fig. 5. Vertical profiles of annual mean CO_2 and seasonal cycle amplitudes for each full year of observations. The annual means for the top measurement level at each tower for each year (given in the key) are subtracted from all observations for that year.

results in which only the amplitude of the seasonal cycle, and not the phase, are shown, such as Fig. 8 of Fung et al. (1987) and Fig. 8 of Denning et al. (1996b).

3.3. Annual means: comparison to "background" sites

The annual mean CO_2 mixing ratios at each level on the NC and WI towers are shown in Fig. 5. For each year the annual mean at the top measurement height (496 m at NC, 396 m at WI, given in the figure key) has been subtracted from all the data to facilitate comparison between years. At each tower there is a remarkable year-to-year consistency in the vertical gradient of the annual means indicating that annual means are set by regional and continental scale processes rather than by local effects such as changes in vertical mixing or local biospheric exchange.

Fig. 6 shows annual mean CO_2 mixing ratios for 1995 from flask data obtained by the CMDL cooperative air sampling network (Conway et al., 1994). Differences between the annual mean flask data and in situ data for the towers reflect details of the diurnal cycles at the towers that are not captured by the flasks that are grab samples. For example, at the NC tower (ITN) flasks are typic-

ally filled between 1440 and 1640 GMT which is close to the time of the CO_2 maximum at 496 m associated with the onset of vigorous convection (Fig. 1). The flask data from HUN (a 110-m tall tower in Hungary) are no doubt affected by similar considerations and cannot be considered to be representative of 24-h means. Continuous in situ measurements have been carried out at that site since 1994 (Haszpra and Nagy, 1997). Coastal sites such as TAP, BSC, and CMO (see below) could also be affected by diurnal cycles as a result of sea-breeze phenomena, however, continuous sampling has not been carried out at these sites. Most of the other stations of the sampling network have been carefully selected to represent regional average mixing ratios and are not likely to be affected significantly by diurnal effects (Conway et al., 1994).

From Fig. 6 it is clear why three-dimensional model simulations that use only data from the "background" sites may give an approximate balance for North America of a net annual CO_2 sink to the terrestrial biosphere and the total source from fossil fuel combustion (Fan et al., 1998). Sites that bracket North America zonally show very similar annual mean mixing ratios. For example, CBA and SHM in the North Pacific are similar to MHT in the North Atlantic. Also, BMX in the
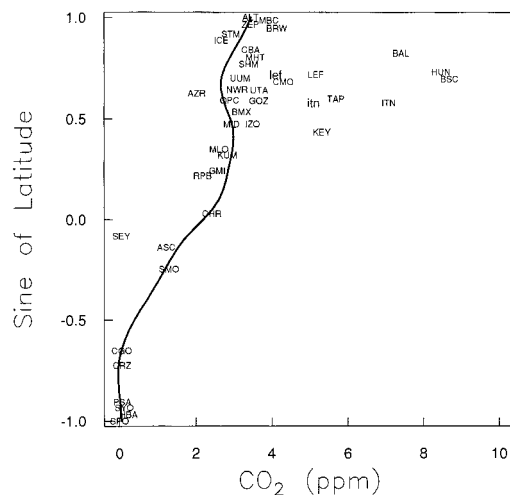
Fig. 6. Annual mean CO_2 mixing ratios for 1995 from flask samples collected approximately weekly at sites of the CMDL cooperative air sampling network plotted as a function of latitude. For some sites the latitudes were shifted slightly to improve legibility. The annual mean for the South Pole (SPO, 357.7 ppm) was subtracted from all data. The three-letter site codes are defined in Tans et al. (1996a), except that we use BMX to represent a combination of the two Bermuda sites (BME and BMW). The solid line is a smoothed representation of the CO_2 mixing ratio in the marine boundary layer from GLOBALVIEW — CO_2 (1997), and includes data from some sites not otherwise shown here. The codes ITN and LEF are flask data from the NC and WI towers, respectively, and the lower case letters for these stations denote annual averages of 24-h mean mixing ratios calculated from the in-situ data.

Atlantic is very similar to the midcontinental mountain top site at NWR and to the MBL reference curve (which is fit to data from BMX and other sites). However, these sites are distant from the continental source/sink regions. Also, oceanic sources and sinks of CO_2 may complicate the interpretation. Several sites that are closer to the continental areas show higher annual mean CO_2 mixing ratios than the MBL curve. These sites include our NC and WI tower sites, the Hungary tower (HUN), a ferry boat in the middle of the Baltic Sea (BAL), a site on the Black Sea coast (BSC) where flasks are filled when wind blows from the sea to the land, the Tae-Ahn Peninsula of South Korea (TAP), and a coastal station in Oregon (CMO).

There are 3 main reasons to expect that annual average CO_2 mixing ratios at the surface in continental locations would be higher than over the oceans, even if the terrestrial biosphere were in balance (zero annual net flux). First, fossil fuel combustion takes place primarily on the land surface. In Subsection 3.4 we examine the extent to which the fossil fuel signal can be accounted for at the NC and WI towers based on measurements of CO. Second, the diurnal covariance in the sign and magnitude of the CO_2 surface flux and the stability of the PBL, as discussed in Subsection 3.1, leads to higher average CO_2 mixing ratios near the surface over the continents (Denning et al., 1996b). As discussed in Subsection 3.1, the daily average CO_2 mixing ratios at the highest tower measurement levels, which are well above the top of the stable nocturnal PBL, probably minimize this bias towards high CO_2 mixing ratios. The effect is probably also minimized at the flask sites, with the exception of HUN, as a result of careful site selection. Third, a similar seasonal covariance exists between surface fluxes and mean PBL depth and convective cloud venting of the PBL: in summer surface heating is stronger than in winter and summertime PBLs tend to be deeper and more rapidly ventilated (Denning et al., 1996b). The latter mechanism leads to larger annual mean CO_2 mixing ratios over the continents than over the oceans because the wintertime respiration signal is diluted into a shallower PBL than the summertime signal of net photosynthesis. For example, Fan et al. (1998) used the NOAA Geophysical Fluid Dynamics Laboratory (GFDL) SKYHI GCM to examine the role of the terrestrial biosphere (represented using the CASA biosphere model, Potter et al., 1993) in the global budget of atmospheric CO_2. In a simulation with purely seasonal surface exchange of CO_2 (annual mean biospheric exchange for each grid point = 0) the annual mean mixing ratios for the altitude of the mid-PBL (740 m) in the grid cells representing the NC and WI towers were 0.7 and 0.4 ppm, respectively, higher than in a run with no biospheric exchange. These annual mean enhancements are solely the result of covariance between seasonally varying surface exchange and PBL depth and venting.

Without observations of PBL depth and convective cloud activity we can say little about the magnitude of the seasonal covariance effect from our data. Such observations are being carried out

at the WI tower site starting in the spring of 1998. Nevertheless, the current data should be useful to help constrain this effect when properly used in dynamic models.

3.4. The CO_2 source from fossil fuel combustion

To estimate the extent to which CO_2 from regional fossil fuel combustion influences the annual mean CO_2 mixing ratios at the towers we utilize our CO data from the in situ GC measurements (Hurst et al., 1997). The GC observations began at both towers in 1995. Though CO is emitted by several types of sources, and is also produced in the atmosphere photochemically by the oxidation of CH_4 and other hydrocarbons, fossil fuel combustion sources are strongly dominant over most of the conterminous United States (Environmental Protection Agency (EPA), 1995). The mean CO/CO_2 emission ratio for fossil fuel combustion in the United States is about 0.023 mole/mole. We obtained this value by dividing the total fossil fuel combustion emissions of CO for the United States in 1994 from EPA (1995) by the fossil fuel source of CO_2 from Andres et al. (1998). Data from our NC tower and other stations (Bakwin et al., 1994, 1995, 1997; Potosnak et al., 1998) indicate that this emission ratio is reasonably accurate.

Daily mean mixing ratios of CO at the towers are shown in Fig. 7 along with MBL reference curves for these latitudes constructed from CMDL flask measurements (Novelli et al., 1992, describe the measurements of CO in the CMDL flasks). At the towers CO enhancements over the MBL are typically about 20–90 ppb (NC, inner $\approx 70\%$ of the data) and 0–60 ppb (WI) and are roughly constant seasonally; apparently the seasonal cycle of CO at the towers is driven primarily by photochemistry (Novelli et al., 1992) which equally affects the MBL sites. For an estimate of the contribution of fossil fuel combustion to CO_2 at the towers relative to the MBL we divide the difference of the CO daily means at the towers from the MBL reference by the CO/CO_2 molar emission ratio of 0.023. The estimates of annual average fossil fuel CO_2 at the towers are in good accord with the simulation of Denning (1994) showing that CO_2 at the NC and WI towers should exceed the MBL reference by 3.1 and 1.8 ppm, respectively, due to fossil fuel alone (Denning, personal communication, 1998). Subtracting these values from the tower CO_2 mixing ratios gives the values shown in Fig. 8, and the results for the annual means for 1996 and 1997 are given in Table 2. With the fossil fuel CO_2 subtracted the annual mean CO_2 mixing ratios at the towers are 0.1–1.1 ppm (one standard deviation uncertainty for the tower means only is 0.2 ppm, estimated by a Monte Carlo method) lower than the MBL reference, which is suggestive of net uptake of CO_2 by the terrestrial biosphere in the regions around the towers.

Our estimate uses data from the highest tower levels only. Because of the large build-up of CO_2 near the surface at night, we probably underestimate the daily mean CO_2 mixing ratios integrated through the depth of the daytime PBL (or nighttime relic layer), roughly 1.5 km. In midsummer the daily mean at 51 m (30 m) on the NC (WI) tower is about 14 ppm (12 ppm) higher than at 496 m (396 m) as a result of the nighttime build-up (Subsection 3.1). If the lower levels are reasonably representative of the nighttime PBL, then we estimate that the effect of the nighttime build-up (typically $\leqslant 150$ m deep) on the 24-h column average through the lower 1.5 km is roughly 1 ppm in midsummer. The nighttime build-up is much less in other seasons, so as an annual average we calculate that our fossil fuel corrected CO_2 mixing ratios at the towers underestimate the 0–1.5 km column mean by about 0.3 ppm.

Table 2. *Carbon dioxide mixing ratios (ppm) at the towers with and without the fossil fuel contribution subtracted as described in the text*

Year	NC	NC-FF[a]	NC MBL[b]	WI	WI-FF[a]	WI MBL[b]
1996	364.7	362.4	362.9	363.8	362.0	363.1
1997	365.9	363.5	363.9	364.5	363.7	363.8

[a] FF = fossil fuel contribution to CO_2 calculated from CO (see text).
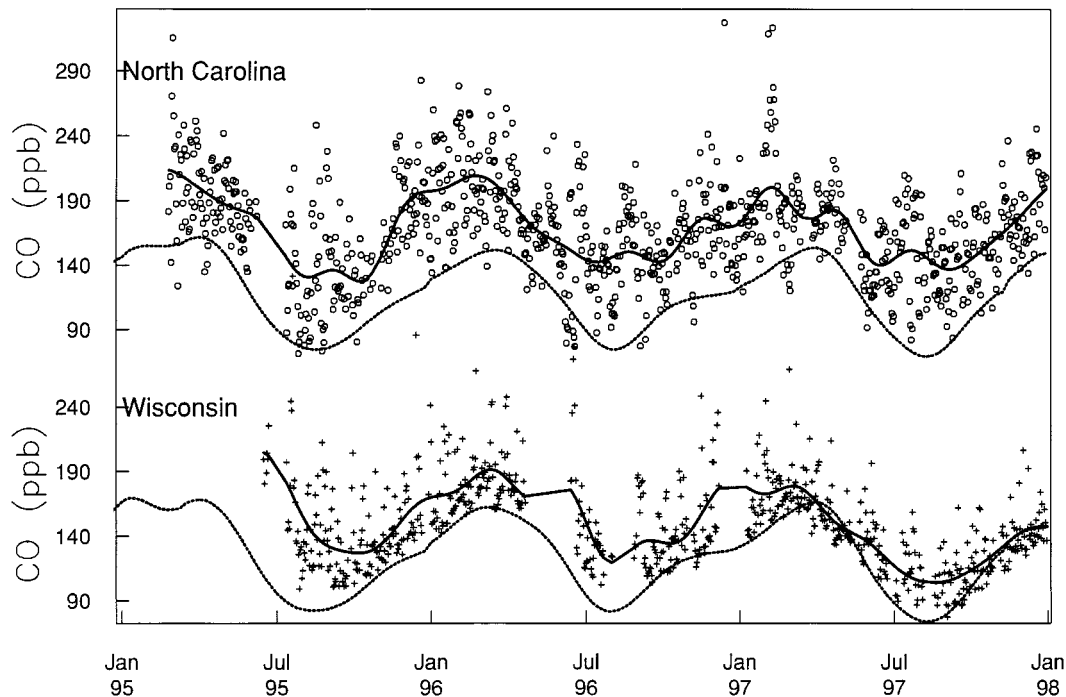[b] MBL = marine boundary layer reference (GLOBALVIEW — CO_2, 1997).

Fig. 7. Daily mean CO mixing ratios at 496 m (NC) and 396 m (WI) above the ground on the towers and the MBL reference curves (dotted) for those latitudes. The tower data are fit with smooth curves as in Fig. 2.

Photochemistry removes CO and the lifetime is about 1.5 months at these latitudes in midsummer, and much longer in other seasons (Novelli et al., 1992). In summer a 10-day average transport time between the continental regions represented by the tower data and the remote MBL would result in loss of about 20 ppb CO leading to an over-estimate of the fossil fuel CO_2 at the towers of about 1 ppm. This bias decreases rapidly away from midsummer and is probably not more than 0.3 ppm as an annual average.

In summary, CO_2 mixing ratios measured at the towers, when corrected for local fossil emissions, appear lower than at MBL sites at the same latitudes. However, we have discussed three possible biases: the seasonal covariance between biological fluxes and PBL mixing which biases the tower CO_2 towards higher annual mean mixing ratios, and the photochemical removal of CO and nighttime build-up of CO_2 near the ground both of which bias the tower CO_2 to lower mixing ratios. Hence, these effects are at least partially offsetting. Detailed photochemical and transport

modeling and observations of PBL depth over a full annual cycle are needed to fully evaluate these biases.

3.5. Data access

Archived data from the NC and WI towers are available by anonymous ftp from ftp.cmdl.noaa.gov (path: ccg/towers). The tower station codes are "itn" (NC) and "lef" (WI), and it is recommended that the user first retrieve and carefully read the "readme" files for these sites. Available online data include hourly (WI) or half-hourly (NC) CO_2 mixing ratios plus meteorological and auxiliary observations. Daily average CO_2 mixing ratios at each level are also provided, as are statistical summaries (by month) of the diurnal cycles of CO_2. Higher time resolution data can be obtained by contacting the lead author. Also, the tower CO_2 data have been incorporated into the GLOBALVIEW dataset which is available via ftp and on CD-ROM (GLOBALVIEW — CO_2, 1997). Flask sample data from the towers and
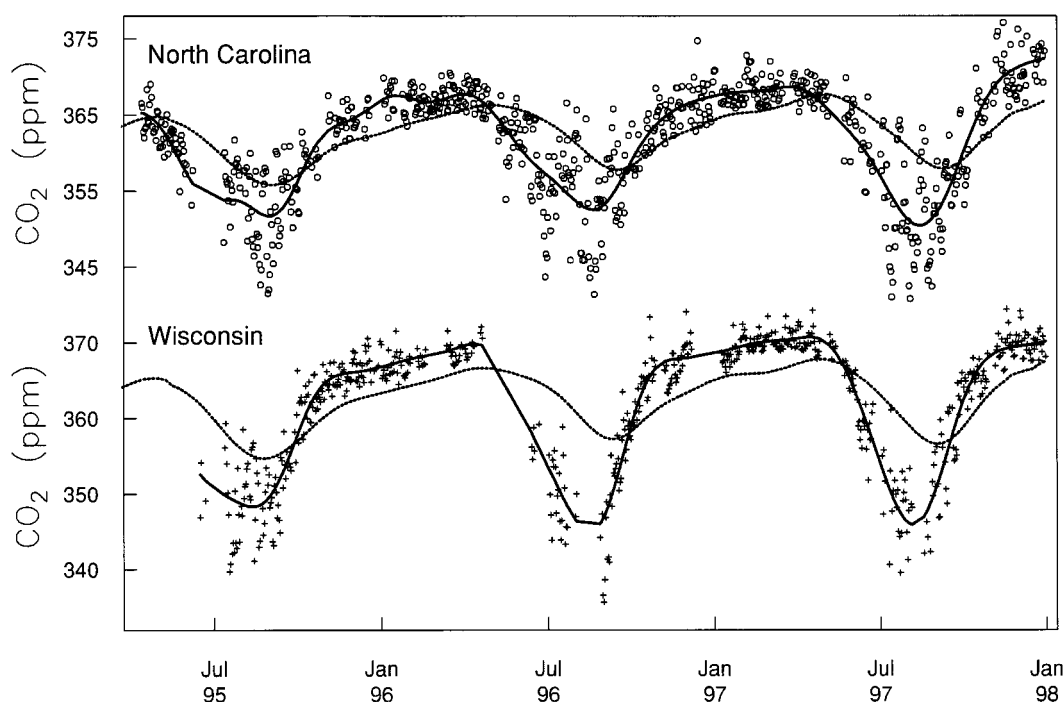
Fig. 8. Daily mean CO_2 mixing ratios at the towers with the contribution from regional fossil fuel combustion removed based on measured CO mixing ratios (see text), and the MBL reference curves (dotted) for those latitudes.

other CMDL stations are also available via ftp from the same server (path: ccg/co2/flask).

4. Summary

We have presented measurements of CO_2 mixing ratios from ongoing observations at two very tall communications towers in the United States. When adjusted for the influence of excess (regional) fossil fuel emissions, the annual average CO_2 at the towers is lower than in the marine boundary layer at the same latitudes indicating that the terrestrial biota in the regions around the towers likely represent a significant net sink for CO_2. Incorporation of the tower data into global and regional models of the carbon cycle will provide a powerful constraint on calculations of the influence of terrestrial biology on atmospheric CO_2. We have described some features of the tower data that will require particular attention when using these data to constrain model simulations, including the diurnal and seasonal cycles of

CO_2 mixing ratio as a function of altitude above the ground, and the seasonal cycle of the short-term variability of CO_2. Successful models will require at least the following characteristics in addition to those already contained in most GCMs: (1) a reasonable parameterization of the diurnal cycle of planetary boundary layer mixing, and (2) a realistic simulation of the diurnal and seasonal cycles of terrestrial biology and the uptake and release of CO_2.

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