# Aircraft and tower measurements of CO<sub>2</sub> concentration in the planetary boundary layer and the lower free troposphere over southern taiga in West Siberia: Long-term records from 2002 to 2011

M. Sasakawa,<sup>1</sup> T. Machida,<sup>1</sup> N. Tsuda,<sup>2</sup> M. Arshinov,<sup>3</sup> D. Davydov,<sup>3</sup> A. Fofonov,<sup>3</sup> and O. Krasnov<sup>3</sup>

Received 29 August 2012; revised 7 August 2013; accepted 8 August 2013.

[1] In situ measurements of the vertical distribution of carbon dioxide  $(CO_2)$  carried out with a light aircraft over a tower site (Berezorechka; 56°08'45"N, 84°19'49"E) in the taiga region of West Siberia from October 2001 to March 2012 document the detailed seasonal and vertical variation of  $CO_2$  concentrations during daytime. The variation appears to be controlled mainly by the  $CO_2$  flux from taiga ecosystems and the height of the planetary boundary layer (PBL). We calculated average CO<sub>2</sub> concentrations in the PBL and the lower free troposphere (LFT), both of which show clear seasonal cycles and an increasing long-term trend. Seasonal amplitude in the PBL had a larger value (29 ppm) than that in the LFT (14 ppm), demonstrating strong CO<sub>2</sub> source-sink forcing by the taiga ecosystems. Mean  $CO_2$  concentrations during 13:00–17:00 local standard time observed at the four levels of the tower (5, 20, 40, and 80 m) showed lower  $CO_2$  concentrations than that observed in the PBL by aircraft during June-August (growing season). This negative bias decreased with increasing inlet height such that the minimum difference appeared at the 80-m inlet ( $-2.4\pm0.8$  ppm). No such bias was observed during other months (dormant season). The daytime CO<sub>2</sub> flux, based on multiple vertical profiles obtained on a single day, ranged from -36.4 to  $3.8 \,\mu\text{mol}\,\text{m}^{-2}\,\text{s}^{-1}$  during July–September. There was a clear difference in the fluxes between the morning and afternoon, suggesting that these data should be considered examples of fluxes during several daytime hours from the West Siberian taiga.

**Citation:** Sasakawa, M., T. Machida, N. Tsuda, M. Arshinov, D. Davydov, A. Fofonov, and O. Krasnov (2013), Aircraft and tower measurements of CO<sub>2</sub> concentration in the planetary boundary layer and the lower free troposphere over southern taiga in West Siberia: Long-term records from 2002 to 2011, *J. Geophys. Res. Atmos.*, *118*, doi:10.1002/jgrd.50755.

## 1. Introduction

[2] Carbon dioxide (CO<sub>2</sub>) is a major greenhouse gas, and its atmospheric concentration has been systematically monitored for years at many ground-based sites all over the world. The terrestrial biosphere is an important carbon reservoir that controls much of the observed variability of atmospheric CO<sub>2</sub>, including its seasonal cycles and interannual variations [*Arneth et al.*, 2010]. Northern high-latitude ecosystems are thought to be a significant sink of anthropogenic CO<sub>2</sub> emissions, but the magnitude and distribution of this carbon sink are still uncertain [e.g., *McGuire et al.*, 2009; *Hayes et al.*, 2011]. Northern high-latitude regions are particularly sensitive

©2013. American Geophysical Union. All Rights Reserved. 2169-897X/13/10.1002/jgrd.50755

to climate variations and are expected to be greatly influenced by future global warming [e.g., *Zhuang et al.*, 2006; *Intergovernmental Panel on Climate Change (IPCC)*, 2007; *McGuire et al.*, 2009]. Siberia, in northern Eurasia, contains large quantities of plant biomass and soil organic carbon, making it one of the largest carbon reservoirs in the world [e.g., *Houghton et al.*, 2007; *Tarnocai et al.*, 2009; *Kurganova et al.*, 2010; *Schepaschenko et al.*, 2011]. Accurate estimates of carbon fluxes in Siberia are therefore essential both for understanding global and regional carbon cycles and for predicting future changes in the Siberian carbon cycle.

[3] Inverse modeling using atmospheric transport models and atmospheric  $CO_2$  observations can be effective for estimating regional and global carbon fluxes from limited atmospheric observations, and this approach has been successful in deriving reasonable carbon fluxes for most land and ocean areas [e.g., *Chevallier et al.*, 2010; *Bruhwiler et al.*, 2011]. However, few inverse modeling studies have focused on Siberia [*Quegan et al.*, 2011] because the available observations there are sparse relative to its large area. *Chevallier et al.* [2010] argued that extending the observational network into eastern Europe and Siberia is important to reduce uncertainty in fluxes estimated by inversion methods over these regions.

Additional supporting information may be found in the online version of this article.

<sup>&</sup>lt;sup>1</sup>Center for Global Environmental Research, National Institute for Environmental Studies, Tsukuba, Japan.

<sup>&</sup>lt;sup>2</sup>Global Environmental Forum, Tokyo, Japan.

<sup>&</sup>lt;sup>3</sup>V.E. Zuev Institute of Atmospheric Optics, Russian Academy of Sciences, Tomsk, Russia.

Corresponding author: M. Sasakawa, CGER, NIES, 16-2 Onogawa, Tsukuba 305-8506, Japan. (sasakawa.motoki@nies.go.jp)



**Figure 1.** Diurnal variation in CO<sub>2</sub> vertical profiles observed by aircraft (upper panels) on (a) 15 July 2003, (b) 4 August 2003, (c) 9 August 2004, and (d) 10 August 2004. Horizontal bars indicate the top of the PBL on each flight. The arrow on the last flight in Figure 1b means that PBL height was higher than the observed maximum height. Temporal variations in CO<sub>2</sub> concentration observed at four different heights on the BRZ tower are shown as colored circles (lower panels). Open circles indicate mean CO<sub>2</sub> concentration in the PBL observed by the aircraft, and the error bars are  $\pm 1$  SD (see section 3.1).

[4] Periodic measurements were carried out by means of a research aircraft at altitudes of up to 4000 m over Zotino (60°48'N, 89°21'E) in central Siberia at 12- to 21-day intervals [*Levin et al.*, 2002; *Lloyd et al.*, 2002] from 1998 to 2005 by the Max Planck Institute for Biogeochemistry. Continuous CO<sub>2</sub> measurements began at the Zotino Tall Tower Observatory (ZOTTO) in April 2009, and *Winderlich* 

*et al.* [2010] reported large seasonal amplitudes of CO<sub>2</sub>, larger than those observed at continental tall towers under oceanic influence or at tower sites in mountainous areas. Others have carried out campaign measurements in Siberia. For example, *Nakazawa et al.* [1997] measured tropospheric concentrations of CO<sub>2</sub> and trace gases in aircraft campaigns for several years, though only during summer; YAK-AEROSIB

(Airborne Extensive Regional Observations in Siberia) aircraft campaigns in 2006 and 2007 precisely measured the variability of CO<sub>2</sub>, carbon monoxide, and ozone [*Paris et al.*, 2008, 2010]; and the ongoing TROICA project (Trans-Siberian Observations Into the Chemistry of the Atmosphere) has measured CO<sub>2</sub> and other species such as carbon compounds, ozone, nitrogen oxides, and aerosols about once per year along the route of the Trans-Siberian Railroad from Moscow to Khabarovsk since 1995 [e.g., *Turnbull et al.*, 2009].

[5] Despite these efforts, the available  $CO_2$  observations remain too sparse to fully constrain carbon fluxes in Siberia with inverse modeling. To overcome this problem and to capture seasonal cycles, vertical profiles, and long-term trends, the Center for Global Environmental Research (CGER) of the National Institute for Environmental Studies (NIES) of Japan, with the cooperation of the Russian Academy of Science (RAS), began periodic flask sampling for greenhouse gas measurements from aircraft over three sites in Siberia in 1993. In addition, in 2001, CGER/NIES and RAS began establishing the Siberian tower network JR-STATION (Japan-Russia Siberian Tall Tower Inland Observation Network), which at present consists of nine towers, to observe regional and short-term variations of greenhouse gases (CO<sub>2</sub> and CH<sub>4</sub>) and to produce data for inverse modeling to obtain regional carbon estimates [Watai et al., 2010; Sasakawa et al., 2010, 2012]. At the JR-STATION site, Berezorechka (BRZ), in West Siberia, a light aircraft measures vertical profiles of CO<sub>2</sub> from the planetary boundary layer (PBL) to the lower free troposphere (LFT).

[6] In this paper, we evaluate the temporal variation of  $CO_2$  concentration in the PBL and the LFT using measurements carried out on and above the tower at BRZ from 2001 to 2011. The parallel tower and airborne measurements also offer insight into how data acquired from a tower or by flask sampling at ground level can represent the mean  $CO_2$  concentration in the PBL.

# 2. Methods

# 2.1. Tower Observations

[7] We focused on the JR-STATION site with the longest record, beginning in 2002, Berezorechka tower (56°08'45" N, 84°19'49"E), which is located in a homogeneous landscape of predominantly taiga (supporting information). We installed a freight container equipped with a gas analyzer and a data logger at the base of the tower. Atmospheric air was sampled from the tower at levels 5, 20, 40, and 80 m above the ground. Sampled air was dried and then introduced into a nondispersive infrared analyzer (NDIR) (LI-820, LI-COR USA; LI-7000 was used before September 2008). The air-sampling flow path was rotated every 12 min (every 6 min before May 2006); that is, the 80 m inlet was sampled on the hour at hh:00, the 40 m inlet at hh:12, the 20 m inlet at hh:24, the 5 m inlet at hh:36, and a reference gas at hh:48. For each 12 min sampling period, air was pumped continuously through the sample line from the inlets, a glass water trap, a semipermeable membrane dryer (PD-625-24SS, Permapure, USA), a stainless-steel tube containing chemical desiccant (magnesium perchlorate), and the NDIR cell. The data produced by the sensors during the last 2 min (1 min before May 2006) were averaged and taken as the representative data for the applicable 1 h period. Thus, all data from the four inlets are shown at the nominal time hh:30 (hh:00 and hh:30 before May 2006). Three standard gases were prepared from pure CO<sub>2</sub> diluted with purified air, and their concentrations were determined against the NIES 09 CO<sub>2</sub> scale [*Machida et al.*, 2011]. Twice a day, the three standard gases were analyzed over the course of an hour, and the signal baseline drifts of the standard gases in 11 h were estimated from the temporal variation of the reference gas signals [*Watai et al.*, 2010]. Measurement precision was  $\pm 0.3$  ppm for CO<sub>2</sub> [*Watai et al.*, 2010; *Sasakawa et al.*, 2010, 2012].

[8] The daily minimum  $CO_2$  concentration at the tower was generally observed in the afternoon when active vertical mixing occurred in the PBL (Figure 1). Therefore, we calculated daytime means by averaging the data observed during 13:00–17:00 local standard time (LST) to represent means for the wider region [*Watai et al.*, 2010]. The daytime mean was calculated only when more than four data were obtained during 13:00–17:00 LST. Furthermore, for the daytime mean calculation, we used only data with a variability (maximum minus minimum) of less than 10 ppm. We adopted this criterion to exclude temporary variations and the influence of sporadic local events that occurred despite there being almost no local anthropogenic emissions.

#### 2.2. Aircraft Observations

[9] A small CO<sub>2</sub> measurement device based on an NDIR (LI-800, LI-COR, USA) equipped with flow and pressure regulation system was developed and installed in a single-engine biplane utility aircraft (Antonov An-2) with a 1000 hp radial engine. The measurement device was similar to the continuous CO<sub>2</sub> measuring equipment used by commercial airliners [Machida et al., 2008]. A sample inlet was set between the two port wings to avoid contamination from the engine exhaust located behind the engine on the starboard side. Atmospheric air was delivered via a Decabon tube into the NDIR after drying with magnesium perchlorate. Two standard gases determined against the NIES 09 CO<sub>2</sub> scale [Machida et al., 2011] were introduced into the NDIR every 5 min. The system used signal averaging to obtain a precision  $(1 \sigma \text{ in } 2 \text{ s}) \text{ of } \pm 0.3 \text{ ppm}$ . Air temperature and relative humidity were measured with a sensor (HMP45A, Vaisala, Finland) fixed next to the inlet. The aircraft ascended up to 2 km (winter) or 3 km (summer) above the ground and then descended to approximately 0.1 km altitude in less than 30 min, yielding a vertical profile of CO<sub>2</sub> concentration. These aircraft measurements were usually conducted in the afternoon on days of good weather, with a frequency of one to four times per month from October 2001 to December 2008 and once per several months after that. We also conducted multiflight measurements (several flights in one day) to capture diurnal variations in the vertical  $CO_2$  profile (20 days). The data are available from http://db.cger.nies.go. jp/ged/data/Siberia/aircraft/CO2/in-situ/.

#### 2.2.1. Mean Concentration Within the PBL and LFT

[10] To compare the temporal CO<sub>2</sub> variation between the PBL and the LFT, we determined the height of the PBL at the time of each flight from the level of the maximum vertical gradient of potential temperature, indicative of a transition from a convectively less stable region below to a more stable region above. The level of the minimum vertical gradient of

 Table 1. Number of Days in Each Season That PBL Height Was

 Determined by Each Method

	Method Used for Determining PBL Height								
Season	Potential Temperature	Specific Humidity	Surface-Based Inversion	Higher Altitude <sup>a</sup>					
DJF	29	4	13	0					
MAM	28	14	2	13					
JJA	28	21	0	27					
SON	34	9	6	4					

<sup>a</sup>An altitude higher than the maximum observation level.

specific humidity was also used to determine the PBL height. These methods for estimating PBL height are based on the methods reviewed by Seidel et al. [2010]. When both meteorological data were obtained, the PBL height that better agreed with the transition in CO<sub>2</sub> concentration was adopted. The top of a surface-based inversion was often adopted as the PBL height during winter. While the potential temperature and specific humidity methods both allow for the possibility of an unstable or neutral PBL, a surface-based inversion is a clear indicator of a stable boundary layer whose top can define the PBL height. The data streams for potential temperature and specific humidity (2 s sampling time) were fitted with spline curves that were used for the gradient calculation. When we could not determine the top of the PBL with the above methods, we set the top at an altitude higher than the maximum observation level. The number of days in each season that PBL height was determined by each method is shown in Table 1.

[11] We derived average aircraft data for every 50 m increment of altitude, using the observed  $CO_2$  data within 50 m of that altitude with a temporal resolution of 2 s, to avoid unequal weighting of the different atmospheric layers. For example, average data for the 1000 m level were produced from the data obtained between 950 and 1050 m. If no data were obtained in a given 100 m increment, as in the case of two standard gas measurements, the average for that altitude was determined from the average data from adjacent levels. Some flight observations ended during two standard measurements, which caused no data to be recorded near the ground surface. In that case, the average was taken from the nearest level above. For example, when there were no data between 100 and 200 m altitude, we repeated the value from the 200 m level at the 150 m level.

[12] We then obtained  $CO_2$  averages for the PBL from the series of 50 m bin data below the PBL height. We also calculated  $CO_2$  averages in the LFT from the bins above the PBL height. We excluded the 50 m bin that included the top of the PBL from both calculations to exclude the transition layer where  $CO_2$  concentration changed sharply. The  $CO_2$  concentration in the LFT was relatively stable, but it sometimes varied considerably just above the transition layer. In those cases, if a stable layer with small  $CO_2$  variation was documented above the transition layer, the average for the LFT was calculated from the stable layer.

#### 2.2.2. Estimates of the Large-Scale CO<sub>2</sub> Flux

[13] When vegetation is actively growing and boundary layer conditions are convective, plant assimilation uptake and the  $CO_2$  exchange between the PBL and the LFT drive the diurnal  $CO_2$  evolution in the PBL. As shown by *Culf et al.* [1997] and *Pino et al.* [2012], a simple box model can account for the evolution of the  $CO_2$  concentration (Figure 2). At each time step (the flight interval was used in these calculations), the mass of  $CO_2$  in the PBL is increased by the addition of the masses of  $CO_2$  introduced into the PBL from below (surface flux) and from above (owing to the known increase in height of the PBL). The new concentration in the PBL is then this total mass of  $CO_2$  spread over the new measured height of the PBL.

[14] Differences in molar density at different times can be equated to the average surface flux over a time period as follows:

$$\langle F \rangle = \frac{1}{T} \sum_{i=1}^{h_{n+1}} \left[ \rho_{n+1}(i) C_{n+1}(i) - \rho_n(i) C_n(i) \right] \Delta z, \tag{1}$$

where T is the integration period from time  $t_n$  to  $t_{n+1}$ , and  $\rho$  is the molar air density  $(mol m^{-3})$ . This method is modified from the version used by Wofsy et al. [1988] to infer regional CO<sub>2</sub> fluxes over the Amazon basin. Although Wofsy et al. [1988] integrated the column abundance of  $CO_2$  from the ground surface to an arbitrary height (horizontal dotted line in Figures 2a and 2c), we calculated it up to the PBL height  $h_{n+1}$  at time  $t_{n+1}$  (shaded area in Figures 2a and 2c). This change reduces the potential error derived from the CO<sub>2</sub> variation in the LFT. Although the variation in the  $CO_2$ concentration in the LFT was relatively small in most cases, we did not use data in which the CO<sub>2</sub> concentration in the LFT decreased by more than the measurement precision (0.3 ppm) between consecutive flights. The average CO<sub>2</sub> difference between consecutive flights in the LFT was  $\pm 1.2 \pm 1.2$  ppm (mean  $\pm$  SD) (n = 11). Thus, the calculated rate of CO<sub>2</sub> uptake should be a conservative estimate.

[15] The continuous molar CO<sub>2</sub> density data ( $\rho$ C) were averaged over every 50 m increment of altitude by the method described in section 2.2.1. The 50 m bin data were used for the flux calculation with  $\Delta z$  being 50 m. We chose



**Figure 2.** The structure of the mixed layer model (modified from *Culf et al.* [1997]): (a) at time  $t_n$  (flight *n* at the day); (b) the components making up the composition of the PBL at time  $t_{n+1}$  (flight n+1 at the day); (c) the structure at time  $t_{n+1}$ . C<sub>LFT</sub> and *C*(*i*) are the CO<sub>2</sub> concentration in the LFT and in the layer *i*, respectively; *i* represents the layer from  $z_{i-1}$  to  $z_i$ , and  $\Delta z = z_i - z_{i-1}$ ; *h* is the PBL height; and *F* is the average flux of CO<sub>2</sub> into the PBL at the surface between  $t_n$  and  $t_{n+1}$ .



**Figure 3.** (top) Temporal variations in  $CO_2$  vertical profiles observed by flights during 2001–2012 and (bottom) variations in the PBL height calculated from observed meteorological data from each flight. Blue dots indicate the top of a surface-based inversion, which was defined as the PBL height. Red dots indicate that the PBL height was higher than the observed height (see section 2.2.1).

consecutive flights in the normal daytime situation of a growing PBL, rather than a shrinking PBL, to minimize any error due to exchanges of  $CO_2$  with air above the top of the PBL. For  $t_n$  and  $t_{n+1}$ , we adopted the times at each minimum height. We also calculated the flux with  $h_{n+1} \pm 50$  m to check the sensitivity of the flux to the determined PBL height.

[16] *Yi et al.* [2000] showed that the contribution of horizontal advection to the net ecosystem-atmosphere exchange of CO<sub>2</sub> (NEE) could be considerable in a heterogeneous landscape. However, *Yi et al.* [2000] also mentioned that NEE can be obtained by the method of *Wofsy et al.* [1988] if CO<sub>2</sub> sources and sinks are homogeneous and the terrain is flat. The BRZ tower is located in a homogeneous, predominantly taiga landscape (supporting information); hence, CO<sub>2</sub> sources and sinks should be homogeneous. We therefore assumed that the contribution of horizontal advection was negligible. Furthermore, to minimize the effect of horizontal advection, we compared footprints calculated with the Stochastic Time Inverted Lagrangian Transport (STILT) model between  $t_n$  and  $t_{n+1}$  (see section 2.3) and excluded the result if the footprints differed.

#### 2.3. STILT Atmospheric Transport Model

[17] We used the STILT model to simulate the transport of air parcels backward in time from the receptor point (BRZ tower). STILT calculates back trajectories using meteorological wind fields while releasing ensembles of model

particles that represent the air parcel. Analyzed wind fields are interpolated to the location of each particle, and the particles themselves are subject to stochastic perturbations that are parameterized to represent the effects of turbulent transport. The density of particles is used to calculate the footprint [Lin et al., 2003; Lin et al., 2004]. The footprint, in units of ppm/( $\mu$ mol m<sup>-2</sup> s<sup>-1</sup>), provides the linkage between concentrations and surface fluxes and represents the sensitivity of the atmospheric concentration (ppm) at the starting location to upstream fluxes ( $\mu$ mol m<sup>-2</sup> s<sup>-1</sup>). This means that in the backward-time run, emissions upstream of the receptor at a location with higher particle density have a greater contribution to changes in the mixing ratio at the receptor. STILT was driven with reanalysis data from the National Centers for Environmental Prediction and the National Center for Atmospheric Research, which have a temporal resolution of 6 h and a spatial resolution of  $2.5^{\circ} \times 2.5^{\circ}$ . The model, which was run backward in time for 3 days, provided surface influence functions with a grid of  $1^{\circ} \times 1^{\circ}$  resolution.

#### 3. Results and Discussion

#### 3.1. Diurnal Flight Observations During Summer

[18] Figure 1 compares diurnal variation in vertical  $CO_2$  profiles observed in the multiflight measurements (upper panels) and continuous data observed at the BRZ tower (lower panels) during summer. The top of the PBL, shown



**Figure 4.** Temporal variation in mean  $CO_2$  concentrations in the PBL (blue circles) and LFT (black circles) from aircraft observations. Fitted curves (solid lines) and trend lines (dashed lines) are also shown.

in the upper panels of Figure 1, rose rapidly in the morning and remained high until the evening. This daytime growth in the PBL was observed during most flights. Our data captured the diurnal variation in PBL height with its maximum during the afternoon, although the timing and maximum height varied from day to day. Anomalously high CO<sub>2</sub> concentrations were sometimes observed at low altitudes in the early morning when the boundary layer was stable; these represented CO<sub>2</sub> accumulation from nocturnal respiration by the taiga ecosystems. These anomalies disappeared, and a clear difference in CO<sub>2</sub> concentration between the PBL and the LFT was consistently observed after the PBL developed because photosynthesis started after sunrise, and then the lower-CO<sub>2</sub> air from the taiga was elevated to the top of the PBL. The vertical CO<sub>2</sub> distribution above the stable boundary layer in the early morning depends on the concentration in the residual layer from the day before; thus, it showed unique profiles independent of the PBL height. The CO<sub>2</sub> concentration in the PBL always decreased from morning to afternoon, which may reflect continuous  $CO_2$  absorption by the taiga vegetation. On the other hand,  $CO_2$  concentration above the PBL (in the free troposphere) did not show regular trends within the day and may have been influenced by long-range transport.

[19] To compare the variations in  $CO_2$  concentration observed by aircraft within the PBL and those observed from the BRZ tower, Figure 1 displays the  $CO_2$  average in the PBL (see section 2.2.1) together with the tower data. Diurnal variation in average  $CO_2$  concentrations in the PBL in aircraft data agreed well with tower data after the development of the PBL, that is, except during early morning (lower panels of Figure 1). Although the data from the BRZ tower basically exhibited similar variations, the data from lower inlets showed lower concentrations, which suggests strong  $CO_2$  absorption due to photosynthesis at the surface during the daytime. When  $CO_2$  emission due to respiration exceeded  $CO_2$  absorption due to photosynthesis, the concentration gradient reversed as shown in the early morning and evening.

#### 3.2. Routine Flight Observations

[20] Vertical profiles of  $CO_2$  concentration observed by 275 routine flights from October 2001 to March 2012 are shown in Figure 3. On the 20 days of multiflight measurements, the profile with the greatest PBL height was chosen.

Concentrations of  $CO_2$  clearly decreased near the ground during summer, consistent with photosynthesis by the taiga vegetation during the growing season. In contrast,  $CO_2$ concentrations were higher at lower altitudes during the dormant season.

[21] The daytime PBL is pronounced in inland continental locations such as Siberia, varying in thickness from 200–600 m in winter to as much as 2800 m in summer [*Lloyd et al.*, 2002]. We documented clear seasonal variations in PBL height although it sometimes exceeded the height of observations, particularly during summer (Figure 3). The PBL height reached over 3 km during summer as a result of strong convection from solar heating of the ground. During winter, temperatures sometimes decreased below  $-30^{\circ}$ C, and surface-based inversions were common. This condition suppressed the development of the PBL, which then accumulated CO<sub>2</sub> from respiration by the taiga ecosystems.

[22] We fitted curves to the average  $CO_2$  concentrations in the PBL and LFT using the digital filtering technique of Thoning et al. [1989]. Both fitted curves exhibited a longterm increasing trend with a clear seasonal cycle (Figure 4). The average seasonal cycle for the PBL (Figure 5) was characterized by a minimum in late July and a maximum in November-December, yielding a seasonal amplitude of 29 ppm. This amplitude was about twice the one observed in the LFT (14 ppm), and the seasonal minimum in the PBL occurred about half a month earlier than the minimum in the LFT, demonstrating the strong CO<sub>2</sub> source-sink forcing by the taiga ecosystem. Similarly, Llovd et al. [2002] documented a large difference between the PBL and the LFT in the magnitude of the seasonal amplitude (25 and 15 ppm, respectively) over Zotino in central Siberia, calculating seasonal amplitudes from CO2 flask data in 1998-2000 with the same filtering technique we used. They also found that the spring decrease and the autumn increase in the  $CO_2$ concentrations occurred earlier in the PBL than in the free troposphere. Their flask sampling was done at about 80 m above the treetops and at 500, 1000, 1500, 2000, 2500, and 3000 m height. Their data are too sparse to investigate representative CO<sub>2</sub> variations in the PBL, because CO<sub>2</sub> concentrations in the PBL varied widely, particularly at lower altitudes.



**Figure 5.** Seasonal cycle in mean  $CO_2$  concentrations in the PBL (solid line) and LFT (dashed line) from aircraft observations.



**Figure 6.** Seasonal variation of difference in concentration between the daytime mean observed at the 40 m inlet of the BRZ tower and the PBL mean from aircraft observations on the same day. Horizontal solid (dotted) line indicates the mean value during June–August (other months). Data obtained during surface-based inversions are shown in closed symbols and were not used for the average calculation.

The 29 ppm seasonal amplitude from our observations thus likely represents a more realistic value in the PBL.

[23] The trend curve for CO<sub>2</sub> was consistently higher in the PBL than in the LFT (Figure 4), which is mainly thought to be due to a rectifier effect [*Denning et al.*, 1999]. The difference in yearly means (2002–2011) was  $2.0\pm0.5$  ppm. The main physical mechanism of the rectifier effect is the covariation of the biospheric flux and the vertical mixing in the PBL, that is, the simultaneous variation of vertical mixing in the PBL and biospheric flux in which summer photosynthetic uptake and vertical mixing in a deep PBL contrast with winter ecosystem respiratory release and vertical mixing in a shallow PBL. Particularly at BRZ, both the high biospheric flux of the Siberian taiga ecosystems and pronounced seasonal variation in PBL height (Figure 3) might have induced a distinct rectifier effect.

### 3.3. Comparison of Flight and Tower Observations

[24] To ascertain how well the measured concentrations from the tower represented the  $CO_2$  variation in the PBL, we compared the mean CO<sub>2</sub> concentration in the PBL obtained by aircraft with the daytime mean CO<sub>2</sub> concentration obtained from the tower on the same day. The tower means were often lower than the PBL means during June-August, which suggests that tower measurements more directly reflect influences from the ground surface; in this case, the difference was due to a lowering of  $CO_2$  by photosynthesis during the growing season (Figure 6). The bias gradually decreased with increasing height, and the bias in data from the 80 m inlet showed the smallest value of  $-2.4\pm0.8$  ppm (Table 2). On the other hand, the difference in mean concentration between the tower and aircraft data during months other than June-August exhibited no bias regardless of inlet height. This fact indicates that the averaged data from 13:00-17:00 LST are representative of CO<sub>2</sub> variations in the PBL during the dormant season. The data obtained when surface-based inversions occurred were not used for calculating averages because the stable boundary layer would lead to accumulation of CO<sub>2</sub> from respiration by

the taiga ecosystems at lower altitudes. These results suggest that at inland observation sites, it is necessary to evaluate biases in tower-based measurement data, particularly during the growing season.

# 3.4. Carbon Dioxide Flux from Taiga Ecosystems at BRZ

[25] Estimates of the CO<sub>2</sub> flux (section 2.2.2) in July, August, and September are shown in Table 3. Almost all of the calculated daytime fluxes were negative, which indicates that CO<sub>2</sub> was being assimilated by the taiga vegetation during the daytime in summer. Other available CO2 flux observations from the Siberian taiga are sparse. Styles et al. [2002] calculated the daytime CO<sub>2</sub> flux during three periods at Zotino in central Siberia from measurements made on five flights during 23-24 July 1998 and found that it ranged from -13.5 to  $-2.8 \,\mu\text{mol}\,\text{m}^{-2}\,\text{s}^{-1}$ . They also showed that CO<sub>2</sub> fluxes measured by the eddy covariance technique were in the same range  $(-7.0 \text{ and } -4.1 \,\mu\text{mol m}^{-2} \,\text{s}^{-1})$  as the flight data obtained during the same time period. The range of our estimated  $CO_2$  fluxes in July was wider (from -36.4 to 3.8  $\mu$ mol m<sup>-2</sup> s<sup>-1</sup>) than the range reported by *Styles et al.* [2002]. The highest values were obtained in the evening, which suggests that CO<sub>2</sub> uptake in the evening in West Siberia can be unexpectedly strong compared with the previous estimates. The implication therefore is that photosynthesis was active into the evening in July, when the strength of solar radiation is near its annual maximum and sunrise and sunset are at around 4:00 LST and 21:00 LST, respectively. The observations of Styles et al. [2002] were made during consecutive flights separated by long intervals (from 5 to 7 h). During these intervals, the atmospheric footprint might have changed, and such changes would introduce large uncertainty into their estimates because they did not conduct a footprint analysis. Furthermore, one of their flights took place at 21:00 (i.e., around sunset), when the photosynthetic rate may have been very low. These differences might account for the considerable difference in the range of values estimated by Styles et al. [2002] and the range of our estimated values.

[26] It is difficult to discuss variations in the estimated fluxes between months (years) owing to the scarcity of data. However, the estimated values for afternoons in July and August tended to be much lower than morning values. Consequently, regarding any of the estimated fluxes as representative of the entire day's flux involves large uncertainty.

**Table 2.** Difference in Mean  $CO_2$  Concentration (ppm) BetweenTower Daytime Data and Aircraft PBL Data Obtained on theSame Day<sup>a</sup>

	Inlet Height (m)							
Period	5	20	40	80				
June–August	$-4.1 \pm 0.6$	$-3.5 \pm 0.6$	$-3.0\pm0.6$	$-2.4 \pm 0.8$				
	(n=22)	( <i>n</i> =20)	(n=23)	( <i>n</i> =17)				
Other months	$0.3 \pm 0.4$	$0.5 \pm 0.4$	$0.6 \pm 0.4$	$(-1.3\pm0.6)^{\rm b}$				
	(n=63)	( <i>n</i> =69)	(n=66)	(n=27)				

<sup>a</sup>Differences are tower measurements minus aircraft measurements. Ranges are standard error, and *n* indicates the number of data. Data obtained during surface-based inversions were not used for the average calculation. <sup>b</sup>Large error is due to the small number of data.

Table 3.	Carbon	Dioxide	Fluxes	Obtained	from	Consecutive	Vertical	$CO_2$	Profiles	and	Their	Flight	Information	as	Well	as
Footprint (	Codes															

	Flight Number 3					_	$Flux (\mu mol m^{-2} s^{-1})$			
Flight Date 18 July 2002		LST (GMT+6)	Observed Height (m)		PBL Height (m)	Footprint Code <sup>a</sup>	$Mean \pm SE$	+50 m <sup>b</sup>	-50 m <sup>b</sup>	
		9:26 9:40	Max Min	2000	1724	2002Jul18_1000_1500				
	4	11:23 11:37	Max Min	2000 161	1815	2002Jul18_1200_1500	$3.8\!\pm\!0.5$	4.3	3.3	
	7	17:19 17:34	Max Min	1983 150	1598	2002Jul18_1700_1500				
	8	18:44 19:03	Max Min	1999 150	1837	2002Jul18_1900_1500	$-32.6 \pm 0.6$	-33.4	-31.7	
19 July 2002	2	7:20 7:39	Max Min	1933 151	348	2002Jul19_0800_300				
	3	9:20 9:37	Max Min	2000 150	1060	2002Jul19_1000_300	$-4.5 \pm 0.4$	-3.5	-5.5	
12 September 2002	2	8:58 9:16	Max Min	2000 150	244	2002Sep12_0900_200				
	3	11:01 11:15	Max Min	2000 150	1397	2002Sep12_1100_200	$-5.5 \pm 0.6$	-5.3	-5.6	
	4	12:59 13:18	Max Min	2000 150	1381	2002Sep12_1300_1000				
	5	15:17 15:36	Max Min	2000 178	1491	2002Sep12_1600_1000	$-4.1 \pm 0.3$	-4.0	-4.1	
15 July 2003	5	16:33 16:57	Max Min	2998 150	1903	2003Jul15_1700_500				
	6	18:31 18:53	Max Min	2967 233	2394	2003Jul15_1900_500	$-36.4 \pm 0.5$	-36.3	-36.1	
3 August 2003	1	8:59 9:25	Max Min	2915 101	403	2003Aug03_0900_300				
	2	11:28 11:52	Max Min	3000 114	1120	2003Aug03_1200_300	$-19.0 \pm 0.4$	-18.1	-19.3	
9 August 2004	4	12:22 12:45	Max Min	2877 130	1438	2004Aug09_1300_1000				
	5	15:51 16:16	Max Min	3000 100	1573	2004Aug09_1600_1000	$-22.9 \pm 0.2$	-23.5	-22.2	
10 August 2004	3	10:37 11:03	Max Min	3010 151	1389	2004Aug10_1100_1000				
	4	12:29 12:57	Max Min	2901 150	1453	2004Aug10_1300_1000	$-12.0 \pm 0.4$	-11.2	-12.8	
	5	15:32 15:58	Max Min	2978 113	1415	2004Aug10_1600_1000				
	6	16:19 16:44	Max Min	3000 102	1774	2004Aug10_1700_1000	$-22.6 \pm 1.1$	-22.2	-23.6	
12 August 2004	2	9:25 9:50	Max Min	2886	213	2004Aug12_1000_100				
	3	10:10 10:36	Max Min	2927 108	1444	2004Aug12_1100_100	$-9.2 \pm 1.1$	-9.0	-7.7	

<sup>a</sup>Supporting information. Footprint code: YYYYMMMDD\_HHMM\_height.

<sup>b</sup>Calculations below 50 m higher (+50 m) and lower (-50 m) from the PBL height.

Therefore, our data should be considered only as examples of observation-based  $CO_2$  fluxes during a limited daytime period from the West Siberian taiga.

#### 4. Conclusions

[27] Ten years of data from frequent in situ measurements of  $CO_2$  vertical profiles, collected by aircraft over the BRZ tower in the taiga of West Siberia, were used to analyze

variations in the vertical profile of  $CO_2$  concentrations. The  $CO_2$  variation depended on the structure of the PBL, being lower during summer and higher during winter in the PBL. Average  $CO_2$  concentrations in the PBL and the LFT showed clear seasonal cycles and a long-term increasing trend for the observation period; however, seasonal amplitude in the PBL (29 ppm) was greater than that in the LFT (14 ppm). Furthermore, the level of the trend line in the PBL was greater by 2.0 ppm than that in the LFT, which we attributed

to the combination of high activity in the Siberian taiga ecosystems and pronounced seasonal variation in the PBL height, interacting to produce a rectifier effect.

[28] This study gave us useful insight into the characteristics of tower data in modeling  $CO_2$  variation in the PBL. Comparing continuous  $CO_2$  data observed at BRZ and average  $CO_2$  levels in the PBL from aircraft observations, we found that averaging tower data in the afternoon hours (13:00–17:00 LST) could produce representative values for the PBL in daytime, even at the lowest inlet height (5 m). We found that a negative bias of -4.1 to -2.7 ppm could occur during June-August (the peak growing season), with the smallest value at the highest inlet (80 m).

[29] We used the diurnal variations of vertical CO<sub>2</sub> profiles measured multiple times on the same day to obtain daytime CO<sub>2</sub> flux during summer. The estimated fluxes ranged from -36.4 to  $3.8 \,\mu\text{mol}\,\text{m}^{-2}\,\text{s}^{-1}$  during July–September. Since the fluxes in the afternoons in July and August tended to be much lower than those in the mornings, these data should be considered to represent examples of observation-based CO<sub>2</sub> fluxes during some daytime hours from the West Siberian taiga.

[30] Estimates of monthly fluxes of  $CO_2$  on a subcontinental scale over Siberia made using the inverse modeling approach with vertical  $CO_2$  data from aircraft observations and continuous  $CO_2$  data from JR-STATION have been published recently [*Saeki et al.*, 2013].

[31] Acknowledgments. We thank Sergey Mitin (Institute of Microbiology, Russian Academy of Sciences) for administrative support and John C. Lin (University of Utah) for help with STILT calculations. This research was supported by the Global Environment Research Account for National Institutes of the Ministry of the Environment, Japan, through its funding of the project "Analysis for long-term variation in greenhouse gases ( $CO_2$  and  $CH_4$ ) using a tower network over Siberia." Some diurnal flights were performed with support from the Russian Foundation for Basic Research (RFBR grant 11-05-00470).

#### References

- Arneth, A., et al. (2010), Terrestrial biogeochemical feedbacks in the climate system, *Nat. Geosci.*, 3, 525–532, doi:10.1038/ngeo905.
- Bruhwiler, L. M. P., A. M. Michalak, and P. P. Tans (2011), Spatial and temporal resolution of carbon flux estimates for 1983-2002, *Biogeosciences*, 8, 1309–1331, doi:10.5194/bg-8-1309-2011.
- Chevallier, F., et al. (2010), CO<sub>2</sub> surface fluxes at grid point scale estimated from a global 21 year reanalysis of atmospheric measurements, *J. Geophys. Res.*, 115, D21307, doi:10.1029/2010JD013887.
- Culf, A. D., G. Fisch, Y. Malhi, and C. A. Nobre (1997), The influence of the atmospheric boundary layer on carbon dioxide concentrations over a tropical forest, *Agric. For. Meteo.*, 85, 149–158.
- Denning, A. S., T. Takahashi, and P. Friedlingstein (1999), Can a strong atmospheric CO<sub>2</sub> rectifier effect be reconciled with a "reasonable" carbon budget?, *Tellus, Ser. B*, *51*, 249–253.
- Hayes, D. J., A. D. McGuire, D. W. Kicklighter, K. R. Gurney, T. J. Burnside, and J. M. Melillo (2011), Is the northern high-latitude land-based CO<sub>2</sub> sink weakening? *Global Biogeochem. Cycles*, 25, GB3018, doi:10.1029/2010GB003813.
- Houghton, R. A., D. Butman, A. G. Bunn, O. N. Krankina, P. Schlesinger, and T. A. Stone (2007), Mapping Russian forest biomass with data from satellites and forest inventories, *Environ. Res. Lett.*, 2, 045032, doi:10.1088/1748-9326/2/4/045032.
- Intergovernmental Panel on Climate Change (2007), Synthesis Report. Contribution of Working Groups I, II and III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by Core Writing Team, R. K. Pachauri, and A. Reisinger, pp., 104, Cambridge University Press, Cambridge.
- Kurganova, I. N., V. N. Kudeyarov, and V. O. L. De Gerenyu (2010), Updated estimate of carbon balance on Russian territory, *Tellus, Ser. B*, 62, 497–505, doi:10.1111/j.1600-0889.2010.00467.x.

- Levin, I., et al. (2002), Three years of trace gas observations over the EuroSiberian domain derived from aircraft sampling: A concerted action, *Tellus, Ser. B*, *54*, 696–712.
- Lin, J. C., C. Gerbig, S. C. Wofsy, A. E. Andrews, B. C. Daube, K. J. Davis, and C. A. Grainger (2003), A near-field tool for simulating the upstream influence of atmospheric observations: The Stochastic Time-Inverted Lagrangian Transport (STILT) model, J. Geophys. Res., 108(D16), 4493, doi:10.1029/2002JD003161.
- Lin, J. C., C. Gerbig, S. C. Wofsy, A. E. Andrews, B. C. Daube, C. A. Grainger, B. B. Stephens, P. S. Bakwin, and D. Y. Hollinger (2004), Measuring fluxes of trace gases at regional scales by Lagrangian observations: Application to the CO<sub>2</sub> Budget and Rectification Airborne (COBRA) study, *J. Geophys. Res.*, 109, D15304, doi:10.1029/ 2004JD004754.
- Lloyd, J., et al. (2002), A trace-gas climatology above Zotino, central Siberia, *Tellus, Ser. B*, 54, 749–767.
- Machida, T., H. Matsueda, Y. Sawa, Y. Nakagawa, K. Hirotani, N. Kondo, K. Goto, T. Nakazawa, K. Ishikawa, and T. Ogawa (2008), Worldwide measurements of atmospheric CO<sub>2</sub> and other trace gas species using commercial airlines, *J. Atmos. Oceanic Tech.*, 25, 1744–1754, doi:10.1175/2008JTECHA1082.1.
- Machida, T., Y. Tohjima, K. Katsumata, and H. Mukai (2011), A new CO<sub>2</sub> calibration scale based on gravimetric one-step dilution cylinders in National Institute for Environmental Studies NIES 09 CO<sub>2</sub> Scale, in *Report of the 15th WMO/IAEA Meeting of Experts on Carbon Dioxide*, Other Greenhouse Gases and Related Tracers Measurement Techniques (ed. Willi A. Brand). Jena, Germany, September 7–10, 2009, WMO/GAW Report No. 194, 165–169.
- McGuire, A. D., L. G. Anderson, T. R. Christensen, S. Dallimore, L. Guo, D. J. Hayes, M. Heimann, T. D. Lorenson, R. W. Macdonald, and N. Roulet (2009), Sensitivity of the carbon cycle in the Arctic to climate change, *Ecol. Monogr.*, 79, 523–555, doi:10.1890/08-2025.1.
- Nakazawa, T., S. Sugawara, G. Inoue, T. Machida, S. Makshyutov, and H. Mukai (1997), Aircraft measurements of the concentrations of CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, and CO and the carbon and oxygen isotopic ratios of CO<sub>2</sub> in the troposphere over Russia, *J. Geophys. Res.*, *102*, 3843–3859.
- Paris, J. -D., et al. (2008), The YAK-AEROSIB transcontinental aircraft campaigns: New insights on the transport of CO<sub>2</sub>, CO and O<sub>3</sub> across Siberia, *Tellus, Ser. B*, 60, 551–568, doi:10.1111/j.1600-0889.2008.00369.x.
- Paris, J. -D., A. Stohl, P. Ciais, P. Nedelec, B. D. Belan, M. Y. Arshinov, and M. Ramonet (2010), Source-receptor relationships for airborne measurements of CO<sub>2</sub>, CO and O<sub>3</sub> above Siberia: A cluster-based approach, *Atmos. Chem. Phys.*, 10, 1671–1687.
- Pino, D., J. V. -G. de Arellano, W. Peters, J. Schroter, C. C. van Heerwaarden, and M. C. Krol (2012), A conceptual framework to quantify the influence of convective boundary layer development on carbon dioxide mixing ratios, *Atmos. Chem. Phys.*, 12, 2969–2985, doi:10.5194/acp-12-2969-2012.
- Quegan, S., C. Beer, A. Shvidenko, I. McCallum, I. C. Handoh, P. Peylin, C. Roedenbeck, W. Lucht, S. Nilsson, and C. Schmullius (2011), Estimating the carbon balance of central Siberia using a landscape-ecosystem approach, atmospheric inversion and Dynamic Global Vegetation Models, *Glob. Change Biol.*, 17, 351–365, doi:10.1111/j.1365-2486.2010.02275.x.
- Saeki, T., et al. (2013), Carbon flux estimation for Siberia by inverse modeling constrained by aircraft and tower CO<sub>2</sub> measurements, *J. Geophys. Res. Atmos.*, 118, 1100–1122, doi:10.1002/jgrd.50127.
- Sasakawa, M., et al. (2010), Continuous measurements of methane from a tower network over Siberia, *Tellus, Ser. B*, 62, 403–416, doi:10.1111/ j.1600-0889.2010.00494.x.
- Sasakawa, M., A. Ito, T. Machida, N. Tsuda, Y. Niwa, D. Davydov, A. Fofonov, and M. Arshinov (2012), Annual variation of CH<sub>4</sub> emissions from the middle taiga in West Siberian Lowland (2005-2009): A case of high CH<sub>4</sub> flux and precipitation rate in the summer of 2007, *Tellus, Ser. B*, 64, doi:10.3402/tellusb.v64i0.17514.
- Schepaschenko, D., I. McCallum, A. Shvidenko, S. Fritz, F. Kraxner, and M. Obersteiner (2011), A new hybrid land cover dataset for Russia: A methodology for integrating statistics, remote sensing and in situ information, J. Land Use Science, 6, 245–259, doi:10.1080/ 1747423X.2010.511681.
- Seidel, D. J., C. O. Ao, and K. Li (2010), Estimating climatological planetary boundary layer heights from radiosonde observations: Comparison of methods and uncertainty analysis, J. Geophys. Res., 115, D16113, doi:10.1029/2009JD013680.
- Styles, J. M., J. Lloyd, D. Zolotoukhine, K. A. Lawton, N. Tchebakova, R. J. Francey, A. Arneth, D. Salamakho, O. Kolle, and E. D. Schulze (2002), Estimates of regional surface carbon dioxide exchange and carbon and oxygen isotope discrimination during photosynthesis from concentration profiles in the atmospheric boundary layer, *Tellus, Ser. B*, 54, 768–783.
- Tarnocai, C., J. G. Canadell, E. A. G. Schuur, P. Kuhry, G. Mazhitova, and S. Zimov (2009), Soil organic carbon pools in the northern circumpolar

permafrost region, *Global Biogeochem. Cycles*, 23, GB2023, doi:10.1029/2008GB003327.

- Thoning, K. W., P. P. Tans, and W. D. Komhyr (1989), Atmospheric carbon dioxide at Mauna Loa Observatory. 2. Analysis of the NOAA GMCC data, 1974-1985, J. Geophys. Res., 94, 8549–8565.
- Turnbull, J. C., et al. (2009), Spatial distribution of  $\Delta^{14}$ CO<sub>2</sub> across Eurasia: Measurements from the TROICA-8 expedition, *Atmos. Chem. Phys.*, 9, 175–187.
- Watai, T., T. Machida, K. Shimoyama, O. Krasnov, M. Yamamoto, and G. Inoue (2010), Development of an atmospheric carbon dioxide standard gas saving system and its application to a measurement at a site in the West Siberian Forest, J. Atmos. Oceanic Tech., 27, 843–855, doi:10.1175/2009JTECHA1265.1.
- Winderlich, J., H. Chen, C. Gerbig, T. Seifert, O. Kolle, J. V. Lavric, C. Kaiser, A. Hoefer, and M. Heimann (2010), Continuous low-maintenance CO<sub>2</sub>/

CH<sub>4</sub>/H<sub>2</sub>O measurements at the Zotino Tall Tower Observatory (ZOTTO) in Central Siberia, *Atmos. Meas. Tech.*, *3*, 1113–1128, doi:10.5194/amt-3-1113-2010.

- Wofsy, S. C., R. C. Harriss, and W. A. Kaplan (1988), Carbon dioxide in the atmosphere over the Amazon Basin, *J. Geophys. Res.*, 93, 1377–1387.
- Yi, C., K. J. Davis, P. S. Bakwin, B. W. Berger, and L. C. Marr (2000), Influence of advection on measurements of the net ecosystem-atmosphere exchange of CO<sub>2</sub> from a very tall tower, *J. Geophys. Res.*, 105, 9,991–9,999, doi:10.1029/2000JD900080.
- Zhuang, Q., J. M. Melillo, M. C. Sarofim, D. W. Kicklighter, A. D. McGuire, B. S. Felzer, A. Sokolov, R. G. Prinn, P. A. Steudler, and S. Hu (2006), CO<sub>2</sub> and CH<sub>4</sub> exchanges between land ecosystems and the atmosphere in northern high latitudes over the 21st century, *Geophys. Res. Lett.*, 33, L17403, doi:10.1029/2006GL026972.