

# CO<sub>2</sub> column-averaged volume mixing ratio derived over Tsukuba from measurements by commercial airlines

M. Araki<sup>1,\*</sup>, I. Morino<sup>1</sup>, T. Machida<sup>1</sup>, Y. Sawa<sup>2</sup>, H. Matsueda<sup>2</sup>, H. Ohyama<sup>3</sup>, T. Yokota<sup>1</sup>, and O. Uchino<sup>1</sup>

<sup>1</sup>National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki 305-8506, Japan

<sup>2</sup>Meteorological Research Institute, 1-1 Nagamine, Tsukuba 305-0052, Japan

<sup>3</sup>Japan Aerospace Exploration Agency, Sengen 2-2-1, Tsukuba 305-8505, Japan

<sup>\*</sup>now at: Department of Chemistry, Faculty of Science Division I, Tokyo University of Science,

1-3 Kagurazaka, Shinjuku-ku, Tokyo 162-8601, Japan

Received: 15 December 2009 – Published in Atmos. Chem. Phys. Discuss.: 9 February 2010 Revised: 4 August 2010 – Accepted: 5 August 2010 – Published: 17 August 2010

Abstract. Column-averaged volume mixing ratios of carbon dioxide  $(X_{CO_2})$  during the period from January 2007 to May 2008 over Tsukuba, Japan, were derived using CO<sub>2</sub> concentrations measured by Continuous CO<sub>2</sub> Measuring Equipment (CME). The CMEs were installed on Japan Airlines Corporation (JAL) commercial airliners, which frequently fly to and from Narita Airport. It was assumed that CO<sub>2</sub> profiles over Tsukuba and Narita are the same. CO<sub>2</sub> profile data for 493 flights on clear-sky days were analyzed in order to calculate  $X_{CO_2}$  with one of two ancillary datasets: "Tsukuba observational" data (rawinsonde and meteorological tower), or "global" forecast/reanalysis and climatological data (NCEP and CIRA-86). The amplitude of the seasonal variation of  $X_{CO_2}$  using the ancillary data measured in Tsukuba ( $X_{CO_2}$ (Tsukuba observational)) was determined by a least squares fit using a harmonic function to roughly evaluate the seasonal variation over Tsukuba. The highest and lowest values of the obtained fitted curve in 2007 for  $X_{CO_2}$  (Tsukuba observational) were 386.4±1.0 and  $381.7 \pm 1.0$  ppm in May and September, respectively, where the errors represent 1 standard deviation of the fit residuals. The dependence of  $X_{CO_2}$  on the type of ancillary dataset was evaluated. The average difference between  $X_{CO_2}$  from global climatological data,  $X_{CO_2}$  (global), and  $X_{CO_2}$  (Tsukuba observational), i.e., the bias of  $X_{CO_2}$  (global) based on  $X_{CO_2}$ 



*Correspondence to:* M. Araki (araki@rs.kagu.tus.ac.jp)

(Tsukuba observational), was found to be -0.621 ppm with a standard deviation of 0.682 ppm. The uncertainty of  $X_{\rm CO_2}$ (global) based on  $X_{\rm CO_2}$  (Tsukuba observational) was estimated to be 0.922 ppm. This small uncertainty relative to the GOSAT precision suggests that calculating  $X_{\rm CO_2}$  using data from airliners and global climatological data can be applied to the validation of GOSAT products for  $X_{\rm CO_2}$  over airports worldwide.

### 1 Introduction

Climate change is one of our most important environmental problems. Over the past 200 years, the concentration of atmospheric carbon dioxide  $(CO_2)$ , a major greenhouse gas, has increased rapidly from about 280 to 380 ppm (IPCC, 2007). This increase in  $CO_2$  concentration enhances radiative forcing of the atmosphere and thus may contribute to climate change. The prediction of future atmospheric CO<sub>2</sub> concentrations and its influence on climate will require accurate quantification of the distribution and variability of CO<sub>2</sub> sources and sinks, which have been derived from atmospheric CO<sub>2</sub> concentration data by using the inversion of atmospheric transport. Atmospheric CO<sub>2</sub> concentrations are measured with high accuracy, with the majority of the measurements being made at ground stations and meteorological towers using flask sampling and/or Non-Dispersive Infra-Red (NDIR) analyzer. However, because of the sparseness of existing ground stations and the limitation of their altitudinal range, present estimates of  $CO_2$  sources and sinks have large uncertainties (Gurney et al., 2002).

Rayner and O'Brien (2001) demonstrated that global space-based observations of monthly mean column-averaged CO<sub>2</sub> volume mixing ratios (VMR; precision  $\leq$ 1%), denoted as  $X_{CO_2}$ , can be useful for reducing the uncertainties in regional (8° × 10° footprint) CO<sub>2</sub> source and sink estimates. Global  $X_{CO_2}$  values can be derived from space-based nadir-looking observations of sunlight scattered from the earth's surface in the near-infrared spectral region (e.g., Mao and Kawa, 2004).

The Greenhouse gases Observing SATellite "IBUKI" (GOSAT) measures the concentrations of CO<sub>2</sub> and methane (CH<sub>4</sub>) from space (Yokota et al., 2004; Hamazaki et al., 2005; Yokota et al., 2009). Global  $X_{CO_2}$  and  $X_{CH_4}$  products are obtained from the Fourier Transform Spectrometer (FTS) of the Thermal and Near-infrared Sensor for Carbon Observation (TANSO) onboard GOSAT.

Although satellite sensors provide global observations, the data are less accurate than ground-based observations and direct air measurements and need to be validated against more accurate independent datasets. Comparisons of total columns of CO, CH<sub>4</sub>, CO<sub>2</sub>, and N<sub>2</sub>O were carried out between the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) satellite instrument and ground-based FTS data obtained from 11 sites (Dils et al., 2006). Values of  $X_{CO_2}$  over Park Falls, Wisconsin, USA, obtained with a ground-based FTS (Bruker IFS 125 HR), were evaluated by an Orbiting Carbon Observatory (OCO) retrieval algorithm and were compared with those obtained with SCIAMACHY (Bösch et al., 2006). The groundbased FTS is a powerful tool for the validation of  $X_{CO_2}$ satellite products. A network of ground-based FTSs that record direct solar spectra in the near-infrared spectral region, named Total Carbon Column Observing Network (TC-CON, Toon et al., 2009), provides essential validation data for SCIAMACHY and GOSAT. The network consists of 14 sites in Europe, Oceania, North America, and Japan as of March 2010. Additional observation sites for the validation of satellite products are expected, e.g., in tropical zones and South America.

The Comprehensive Observation Network for TRace gases by AIrLiner (CONTRAIL) project (Machida et al., 2008) has been observing vertical CO<sub>2</sub> profiles over 43 airports in the world since 2005. Five Japan Airlines Corporation (JAL) commercial airliners are instrumented with the Continuous CO<sub>2</sub> Measuring Equipment (CME) and most of the flights originate from Narita International Airport (hereafter Narita) in Japan. Vertical CO<sub>2</sub> profiles are obtained during ascents and descents of the airliners.  $X_{CO_2}$  values derived from the CONTRAIL profiles can be used to validate the  $X_{CO_2}$  data observed by GOSAT. Tsukuba is the primary validation site for GOSAT, and is situated 40 km from Narita. The CONTRAIL project could dramatically increase

**Table 1.** Types of analyses of  $X_{CO_2}$  for ancillary meteorological and model data.

	Analysis Type <sup>a</sup>			
$X_{\rm CO_2}$ (Type)	$X_{\rm CO_2}({\rm I})$	$X_{\rm CO_2}({\rm I}')$	$X_{\rm CO_2}({\rm II})$	
MRI CO <sub>2</sub> <sup>b</sup> Number density profile NCEP PBL Tropopause	Yes Sonde Yes Sonde	Yes CIRA-86 Yes Sonde	No CIRA-86 No NCEP	

<sup>a</sup> Analysis types are defined by the ancillary meteorological data used to calculate  $X_{CO_2}$  over Tsukuba. Types I and II analyses use Tsukuba observational data and global climatological data, respectively.

<sup>b</sup> In situ CO<sub>2</sub> tower measurements.

the number of GOSAT validation sites. To develop a method of calculating  $X_{CO_2}$  using CONTRAIL data, this paper will focus on using CONTRAIL profiles over Narita with the extensive meteorological data measured in Tsukuba.

In the present work, values of  $X_{CO_2}$  were derived over Tsukuba during the period from January 2007 to May 2008 for which Tsukuba observational data were used to add information at altitudes not measured by the instruments aboard the aircraft. To calculate  $X_{CO_2}$  over other airports around the world, global climatological data must be used in addition because there are limited numbers of nearby observations. Thus two types of datasets were alternatively used as ancillary meteorological data to calculate  $X_{CO_2}$ : Tsukuba observational data (rawinsonde and a meteorological tower) and global climatological data (National Centers for Environmental Prediction [NCEP] and Committee on Space Research [COSPAR] International Reference Atmosphere [CIRA]-86). The dependence of  $X_{CO_2}$  on the type of the dataset was calculated, and the bias and uncertainty of  $X_{\rm CO_2}$ (global) based on  $X_{\rm CO_2}$ (Tsukuba observational) were estimated. Seasonal variation parameters of  $X_{CO_2}$  were obtained by a least squares fit to roughly evaluate the seasonal variation over Tsukuba. In addition, X<sub>CO2</sub>(Tsukuba observational) were compared with  $X_{CO_2}$  obtained by the groundbased FTS (Ohyama et al., 2009) during the study period.

### 2 Analysis

On the present work, data from the Continuous CME in CONTRAIL, which have an overall precision of 0.2 ppm (Machida et. al., 2008), were used to form the majority of the CO<sub>2</sub> profiles in calculating  $X_{CO_2}$ . The data observed during the ascent and descent of the airliners were taken as vertical CO<sub>2</sub> profiles over Narita. Two types of analyses using either Tsukuba observational data (type I analysis) or global climatological data (type II analysis) as the ancillary meteorological data were performed to calculate  $X_{CO_2}$  (namely,  $X_{CO_2}$ (I) and  $X_{CO_2}$ (II)) over Tsukuba. The types of analyses and their nomenclature are summarized in Table 1.



**Fig. 1.**  $CO_2$ -profile assumption over Tsukuba. (a) High-altitude profiles: The solid and dotted lines show cases in which the tropopause is higher and lower, respectively, than the highest observational point by an airliner. The rectangular area is expanded in (b) and (c). (b) Low-altitude profile: The PBL is lower than the lowest observational point. (c) Low-altitude profiles: the solid line shows a case in which the PBL is higher than the lowest observational point. The dotted line shows an example for which the meteorological-tower datum is missing. All type II analyses use a version of the dotted line in (c) for the low-altitude profile. The type I analysis is the case described in the text.

To obtain  $X_{CO_2}$  over Tsukuba, it was assumed that the CO<sub>2</sub> profile over Tsukuba (36.1° N, 140.1° E) is the same as that over Narita (35.8° N, 140.4° E). In addition, assumptions must be made about the CO<sub>2</sub> profiles above and below the altitudes observed by the airliners (from about 0.5–2 km to about 10–11 km). These assumptions are described below.

Measurements have shown that the stratospheric  $CO_2$  concentration is constant above an altitude of about 20 km and lags about five years behind that of the global mean  $CO_2$ concentration in the free troposphere (Aoki et al., 2003). An average of 381.2 ppm in the free troposphere in 2006 with a growth rate of 1.9 ppm/yr (WMO, 2006), for example, yields concentrations of 373.6 and 375.5 ppm in the stratosphere for 2007 and 2008, respectively. When an airliner does not fly above the tropopause, the  $CO_2$  concentration at the highest observational point (altitude) was assumed to be constant up to the tropopause and the profile from the tropopause to 20 km was linearly interpolated with respect to altitude as shown by the solid line in Fig. 1a. When an airliner crosses the tropopause, the profile from the highest observational point to 20 km was assumed to be linear with respect to altitude as shown by a dotted line in Fig. 1a. For a type I analysis, the height of the tropopause was obtained from a rawinsonde observation at the Tateno Aerological Observatory (36.1° N, 140.1° E) of the Japan Meteorological Agency (JMA). The lowest tropopause as identified by the rawinsonde observation was selected from several observed tropopauses, because the lowest tropopause is generally the boundary between the troposphere and the stratosphere. Since the tropopause observations by rawinsondes were made twice a day at 00:00 and 12:00 h UTC, the tropopause height at a given time was determined by linear interpolation. For a type II analysis, the tropopause height was obtained from NCEP's Model Analyses and Forecasts Global Forecast System (GFS), which was usually consistent with the lowest tropopause identified by the rawinsonde observation.

Within the planetary boundary layer (PBL), the CO<sub>2</sub> concentrations observed at a meteorological tower (36.1° N, 140.1° E, Inoue and Matsueda, 1996, 2001) at the Meteorological Research Institute (MRI), Tsukuba, Japan were used in the case of a type I analysis. The CO<sub>2</sub> concentrations were observed at 1.5, 25, 100, and 200 m above the ground with a precision reported to be better than 0.1 ppm using an NDIR analyzer (Inoue and Matsueda, 1996). If the altitude of the lowest observational point of an airliner was higher than that of the PBL, it was assumed that the CO<sub>2</sub> concentration at the lowest observational point (altitude  $0.5 \sim 2 \text{ km}$ ) of an airliner was constant down to the PBL as shown by a solid line in Fig. 1b. If it was lower than the PBL, a linear profile was assumed between the lowest observational point of the airliner and the highest one (200 m) of the meteorological tower as shown by a solid line in Fig. 1c. If the datum from the meteorological tower was missing, the concentration of the lowest observational point of the airliner was extended down to the ground as shown by a dotted line in Fig. 1c (6% of MRI tower data were missing in the present analysis). The PBL height was obtained from NCEP, as systematic determination of the PBL height from rawinsonde data is difficult. The PBL height data from NCEP with a  $1^{\circ} \times 1^{\circ}$  grid were linearly interpolated with respect to the geographical coordinates of Narita. Analyzed PBL heights were available four times (00:00 h, 06:00 h, 12:00 h, and 18:00 h UTC) daily from NCEP, and forecasted PBL heights were used for the midpoint between the analyses. The PBL data were then linearly interpolated to the takeoff or landing times. In a type II analysis, the concentration at the lowest observational point of an airliner was extended down to the ground irrespective of the PBL height, as shown by a dotted line in Fig. 1c.

Rawinsonde data were utilized for the number density profiles of dry-air in type I analyses. The rawinsonde observations were made twice per day at 00:00 h and 12:00 h UTC. The temporally closest rawinsonde data were used. Because the vertical resolution of rawinsonde observations is a few hundred meters, the number density at a specified altitude was obtained by logarithmic interpolation. Observed total densities of air by the rawinsonde were corrected at each altitude for the water number density using the relative humidity observed by the same rawinsonde. Since rawinsonde data exist up to about 30 km, for altitudes over 30 km, the US standard atmosphere, which is a model that defines values for atmospheric temperature, density, pressure, and other properties over a wide range of altitudes (NOAA, NASA, US Air Force, 1976), was used to generate the number density profile. In type II analyses, CIRA-86 data having monthly mean values in a 5°-latitude grid were employed. CIRA-86 data were linearly interpolated from the 5°-latitude grid to the latitude of Narita. Monthly mean values of CIRA-86 were applied on a monthly basis, without daily interpolation.

To obtain  $X_{CO_2}$ , numerical altitudinal integration was executed as a summation of 100-m layers up to an altitude of 85 km. Homogeneously mixed atmosphere was assumed in each layer. Observations by the airliners occurred at intervals of several tens to hundreds of meters. The CO<sub>2</sub> concentration at an altitude of  $100 \cdot n + 50$  m (n = 1, 2, 3...) was linearly interpolated with the two neighboring observational points and the result was utilized as the concentration of the layer from  $100 \cdot n$  to  $100 \cdot (n+1)$  meters. As mentioned in regards to Fig. 1b and c, a linear interpolation between the lowest observational point by an airliner and the highest observational point by the meteorological tower was also performed to calculate the concentrations of the layers for the region with missing data, with an altitudinal width of  $0.5 \sim 2$  km. In the case of the meteorological tower data, the CO2 concentrations of the lowest three layers were estimated as follows:

$$C_{\text{layer}}(0-100) = 0.5 \cdot C_{\text{tower}}(100) + 0.4 \cdot C_{\text{tower}}(25)$$
(1)  
+0.1 \cdot C\_{\text{tower}}(1.5),

$$C_{\text{layer}}(100 - 200) = 0.5 \cdot C_{\text{tower}}(200) + 0.5 \cdot C_{\text{tower}}(100), \quad (2)$$

$$C_{\text{layer}}(200 - 300) = 1.0 \cdot C_{\text{tower}}(200),$$
 (3)

where  $C_{\text{layer}}$  is the CO<sub>2</sub> concentration in the layer and  $C_{\text{tower}}$  is that observed at the tower. Numbers in parentheses for

 $C_{\text{layer}}$  and  $C_{\text{tower}}$  indicate the altitudinal regions (in meters) of the layers and observational heights (in meters) in the tower, respectively. The coefficient of each term in equations (1–3) was obtained based on the assumption that the observed concentrations at 200, 100, 25, and 1.5 m in the tower are the averaged concentrations of the layers between 150 and 300, 50 and 150, 10 and 50, and 0 and 10 m, respectively.

A flight was excluded if its minimum altitude was greater than 4 km or the maximum altitude was less than 5 km because of the large altitudinal range with missing data.

In the present work, using CONTRAIL data for January 2007–May 2008 over Narita and based on the above assumptions, data from 493 flights by 5 airliners in clear-sky were analyzed, since only the clear-sky data are suitable for successful GOSAT retrievals. "Clear-sky" conditions were determined by the solar absorption spectra measured by an FTS in Tsukuba (Ohyama et al., 2009).  $X_{CO_2}$  from the ground level to an altitude of 85 km, i.e., the entire altitudinal range, and  $X_{CO_2}$  for the altitudinal range of 2–10 km (hereafter  $X'_{CO_2}$ ) were calculated and are shown in Fig. 2. A comparison between  $X_{CO_2}$  and  $X'_{CO_2}$  can demonstrate the effect of the low-altitude atmosphere on  $X_{CO_2}$ .

### 3 Results and discussion

## 3.1 Analysis-type dependence for $X_{CO_2}$ : number density of air

To obtain  $X_{CO_2}$  over airports worldwide, global data must be utilized for the number density of air. CIRA-86 is one such global dataset. Meteorological data from rawinsonde measurements, which are convertible to number densities of air, were obtained over Tateno in Tsukuba. In the present work, number densities obtained from CIRA-86 were validated with those calculated from the rawinsonde, since an uncertainty in the number densities increases the error on  $X_{CO_2}$ . Hereafter, a type I analysis that used CIRA-86 data is referred to as a type I' analysis (Table 1). The profiles of CIRA-86 number densities are similar to those of the rawinsonde, and the differences (1–2%) between them in the altitude range of 0–10 km could be from daily pressure variability.

The differences between  $X_{CO_2}$  by CIRA-86 and by rawinsonde were derived for January 2007–May 2008 (Table 2). The average difference between  $X_{CO_2}(I')$  and  $X_{CO_2}(I)$ , which is the bias of  $X_{CO_2}(I')$  based on  $X_{CO_2}(I)$ , was found to be -0.043 ppm with a standard deviation of 0.067 ppm. The uncertainty (={(Bias)<sup>2</sup> + (Standard deviation)<sup>2</sup>}<sup>1/2</sup>) of  $X_{CO_2}(I')$ based on  $X_{CO_2}(I)$  was 0.080 ppm. These were reduced by half for  $X'_{CO_2}$  (Table 2). Because the uncertainties are small, the number densities of CIRA-86 are admissible as global data to derive  $X_{CO_2}$ .



**Fig. 2.** Time series of  $X_{CO_2}$  and  $X'_{CO_2}$  from type I analysis over Narita using CONTRAIL data from January 2007 to May 2008. Data from 493 flights by five airliners were analyzed.  $X_{CO_2}$  (blue marks and solid blue line) were numerically integrated to cover the entire altitudinal range, i.e., from the ground level to the lower thermosphere (85 km), and for  $X'_{CO_2}$  (red marks and dotted red line) over the altitudinal range of 2–10 km. Data on 16 August 2007 were not included in the fit. Daily averaged  $X_{CO_2}$  using the scaling retrieval algorithm by FTS are plotted by green marks.

**Table 2.** Relative uncertainties of  $X_{CO_2}$  based on type I analysis (ppm)<sup>a</sup>. To show small relative values, the uncertainties are given to 3 decimal places.

Altitudinal range		Analysis Type				
			$X_{\rm CO_2}({\rm I})$	$X_{\rm CO_2}({\rm I}')$	$X_{\rm CO_2}({\rm II})$	$X_{\rm CO_2}(\rm II-I')$
$X_{\rm CO_2}$ Entire		Bias	0.0	-0.043	-0.621	-0.578
	Entire	Standard deviation	0.0	0.067	0.682	0.691
		Uncertainty <sup>b</sup>	0.0	0.080	0.922	0.901
		Bias	0.0	-0.018	-0.019	-0.001
$X'_{\rm CO_2}$	2–10 km	Standard deviation	0.0	0.029	0.037	0.022
		Uncertainty <sup>b</sup>	0.0	0.034	0.041	0.022

<sup>a</sup> We assumed that the type I analysis defines the real  $X_{CO_2}$  in the present analysis.

<sup>b</sup> (Uncertainty) = {(Bias)<sup>2</sup> + (Standard deviation)<sup>2</sup>}<sup>1/2</sup>

## **3.2** Analysis-type dependence for $X_{CO_2}$ : Tsukuba observational data and global data

 $CO_2$  ground concentration data are often difficult to obtain for airports worldwide. Therefore the type II analysis does not use the  $CO_2$  ground concentration and assumes that the  $CO_2$  profile is uniform through the PBL, i.e., that there is less influence, compared to the type I analysis, of an air parcel that includes a high concentration  $CO_2$  from a metropolis and/or a local  $CO_2$  source. During the period between January 2007 and May 2008, the differences between  $X_{CO_2}(I)$  and  $X_{CO_2}(II)$  were derived. The average difference, which is the bias of  $X_{CO_2}(II)$  against  $X_{CO_2}(I)$ , was found to be -0.621 ppm with a standard deviation of 0.682 ppm. The uncertainty of  $X_{CO_2}(II)$  based on  $X_{CO_2}(I)$  was 0.922 ppm. In many cases, the observed CO<sub>2</sub> ground concentration was higher than that at the lowest aircraft altitude, which is likely the reason for the bias.

7664

 $X'_{CO_2}$  shows remarkably reduced uncertainties (Table 2), which may indicate small effects caused by CO<sub>2</sub>-profile turbulence around the PBL and small influences by air parcels that include high-concentration CO<sub>2</sub> from the Tokyo metropolitan area and a local CO<sub>2</sub> source. Furthermore,  $X_{\rm CO_2}({\rm I}')$  were compared with  $X_{\rm CO_2}({\rm II})$  in order to subtract the dependence on air particle number-density datasets from the uncertainty of  $X_{CO_2}(II)$  based on  $X_{CO_2}(I)$ . The differences between  $X_{CO_2}(I')$  and  $X_{CO_2}(II)$  were obtained. The average of the differences, which is the bias of  $X_{CO_2}(II)$  against  $X_{\rm CO_2}(I')$ , was found to be -0.578 ppm with a standard deviation of 0.691 ppm. The uncertainty of  $X_{CO_2}$  (II) based on  $X_{CO_2}(I')$  was 0.901 ppm. Although the dependence on the number-density datasets was subtracted, the decrease in the uncertainty of  $X_{CO_2}(II)$  was small. Thus the uncertainty of  $X_{\rm CO_2}({\rm II})$  strongly depends on the profile assumption around the PBL, as Narita and Tsukuba may have inflows of air parcels over the Tokyo metropolitan area and over a local  $CO_2$  source that disturbs the  $CO_2$  profile uniformity around the PBL. In an analysis at another airport that has a uniform  $CO_2$  profile around the PBL, the uncertainty of  $X_{CO_2}$  will be better than that over Tsukuba.

# **3.3** Variability of $X_{CO_2}$ within 6 h around 13:00 h local time (LT): suitability for GOSAT validation

The variability of  $X_{CO_2}$  in a limited time window must be clarified, since regular observations by GOSAT need to be validated by using non-regular observations by the airliners. We focused on a set of profiles in a 6-h window centered on the GOSAT overpass time (around 13:00 h LT). The standard deviations of  $X_{CO_2}$  by type I analyses for windows containing more than 3 flights were determined for the January 2007-May 2008 period. The standard deviations of  $X_{\rm CO_2}$  that were obtained for 14 windows were averaged. Variabilities - the averages of the standard deviations - were found to be 0.52 and 0.42 ppm for  $X_{CO_2}$  and  $X'_{CO_2}$ , respectively. A limited altitudinal range  $(2-10 \text{ km in } X'_{CO_2})$  did not yield a noticeable decrease in variability compared with the full altitudinal range  $(X_{CO_2})$ . The variabilities originated mainly from the variability of the CO<sub>2</sub> profiles observed by airliners. However the variabilities are within the allowance of 1% for GOSAT data retrieval (Yokota et al., 2004). The majority of the uncertainties of  $X_{CO_2}(I' \text{ and } II)$  based on the real  $X_{CO_2}$  can be derived from the difference between the real profiles and the assumed high- and low-altitude profiles and are difficult to estimate. We discuss the uncertainties of  $X_{CO_2}(II \text{ and } I')$  based on  $X_{CO_2}(I)$  in Sects. 3.1 and 3.2. For the validation of GOSAT data, the variability of  $X_{CO_2}(I)$ within 6 h discussed in this section may be one of the most effective uncertainties of  $X_{CO_2}(I)$  based on the real  $X_{CO_2}$ .

### 3.4 Amplitude of seasonal variation

In order to determine the seasonal variation parameters, the  $X_{CO_2}(I)$  data obtained by each flight were fitted to the following function (Matsueda et al., 2008; Thoning et al., 1989):

$$f(t) = a_1 + a_2 \cdot t + a_3 \cdot t^2 + a_4 \cdot \sin(2\pi t) + a_5 \cdot \cos(2\pi t) \quad (4)$$
$$+ a_5 \cdot \sin(4\pi t) + a_7 \cdot \cos(4\pi t),$$

where *t* is a variable of time in the unit of years from the starting date of the computation on 1 January 2003. Coefficient  $a_1$  is the trend of  $X_{CO_2}$  on the starting date;  $a_2$  and  $a_3$  indicate the growth rate and its second order, respectively;  $a_4$  and  $a_5$  describe the first harmonic, i.e., the seasonal cycle; and  $a_6$  and  $a_7$  describe the second harmonic. Since the obtained  $X_{CO_2}$  data in the present work were limited to 15 months, the value of  $a_3$  was fixed at 0.0. The values of  $a_1$ ,  $a_2$ ,  $a_4$ ,  $a_5$ ,  $a_6$ , and  $a_7$  were determined by a least-squares fit. Although Matsueda et al. used the third harmonic, the large variability of  $X_{CO_2}$  in the present work can interfere with the determination of the third harmonic.

Abnormally high concentrations of  $X_{CO_2}$  – several ppm higher than regular data - were measured for a number of flights in August 2007. These may have been caused by inflows of air parcels from over the Tokyo metropolitan area to over Narita or Tsukuba. Flights from and to Narita can be classified into those that take off or land in the northern airspace of the airport, and those that take off or land in the southern airspace. The observational points below an altitude of 1 km were 20-30 km from Tsukuba for flights landing and taking off to the north, and 50-60 km from Tsukuba for those in the south. The high concentrations of  $X_{CO_2}$ of more than 382.5 ppm recorded in August 2007 were almost always from flights that take off or land in the southern airspace. CO<sub>2</sub> profiles with a high concentration of  $X_{CO_2}$ show high concentrations of CO<sub>2</sub> in the low altitudinal region (0-2 km). If the concentration of CO<sub>2</sub> observed in the low altitudinal region by an aircraft that takes off or lands in the southern airspace is higher than that in the MRI tower and is >400 ppm, the implication is that air parcels over the Tokyo metropolitan area that have a high concentration of CO<sub>2</sub> have not reached Tsukuba; thus the assumption of uniformity of the CO<sub>2</sub> profile over Tsukuba and Narita is incorrect. A CO<sub>2</sub> profile with this condition cannot be used for GOSAT data validation. In the present work, only one flight on 16 August 2007 that had a value of  $X_{CO_2}$  of 388.5 ppm had this condition (shown in Fig. 3) and the value of  $X_{CO_2}$ was not included in the fit.

The standard deviation of the differences between  $X_{CO_2}$ and the fitted curve was 0.98 ppm (0.74 ppm for  $X'_{CO_2}$ ). The obtained coefficients are listed in Table 3 and the fitted curves of  $X_{CO_2}$  and  $X'_{CO_2}$  are shown in Fig. 2. Generally in the Northern Hemisphere, plant activity causes the lowest value of  $X_{CO_2}$  to occur around September and the highest around March and April. The timing of the lowest value arises from CO<sub>2</sub> absorption by plant photosynthesis that is sufficiently



**Fig. 3.** An example of a profile of an inflowing air parcel from over the Tokyo metropolitan area to over Narita on 16 August 2007.

larger than CO<sub>2</sub> production by plant respiration, whereas the photosynthetic absorption of CO<sub>2</sub> is less in winter, leading to the highest value. The highest and lowest values of the fitted curve in 2007 were  $386.4 \pm 1.0$  and  $381.7 \pm 1.0$  ppm, respectively, for  $X_{CO_2}$  and  $387.0 \pm 0.8$  and  $381.1 \pm 0.8$  ppm for  $X'_{CO_2}$  in May and September, respectively, where the errors are 1 standard deviation of the residuals in the fit. The amplitude of the tentative seasonal variation, i.e., the peak-to-peak seasonal amplitude, for the 15 months analyzed here was found to be  $4.63 \pm 0.15$  and  $5.91 \pm 0.13$  ppm by the fitted curves for  $X_{CO_2}$  and  $X'_{CO_2}$ , respectively, where the errors are twice the standard deviation of  $a_4$  in the fit.

The values determined for  $a_2$  (2.27±0.14 and 2.45±0.12 ppm/yr, where the errors are 1 standard deviation of  $a_2$  in the fit) show tentative growth rates for  $X_{CO_2}$  and  $X'_{CO_2}$  over Tsukuba during the period from January 2007 to March 2008 that are quite similar to the value of 2.2 ppm/yr for the FTS measurements by Ohyama et al. (2009). Our values agree with the values of other measurements such as the ground-based FTS observation at Park Falls (Wisconsin, USA) from 2004 to 2006 (2 ppm/yr, Yang et al., 2007; Washenfelder et al., 2006) and the SCIAMACHY observations at north-

**Table 3.** Coefficients obtained for least-squares fits with Eq. (4) to  $X_{CO_2}$  over Tsukuba during the period January 2007–May 2008.

Coefficient <sup>a</sup>	Unit	Entire range <sup>b</sup>	2–10 km	
<i>a</i> <sub>1</sub>	ppm	374.1929	373.1888	
$a_2$	ppm/yr	2.2672 <sup>c</sup>	2.4515 <sup>c</sup>	
<i>a</i> <sub>3</sub>	ppm/yr <sup>2</sup>	0.0	0.0	
$a_4$	ppm	2.7184	3.3140	
$a_5$	ppm	0.5133	0.5376	
$a_6$	ppm	-0.3059	-0.4371	
<i>a</i> 7	ppm	0.2523	-0.0222	

<sup>a</sup> Coefficients in Eq. (4). The coefficient  $a_3$  was fixed at 0.0. See text for details.

<sup>b</sup> Entire altitudinal range, i.e., from the ground level to the lower thermosphere (85 km).
<sup>c</sup> One standard deviation of a<sub>2</sub> is 0.1351 for "Entire range" and 0.1175 for "2–10 km".

ern low- and mid-latitudes from 2003 to 2006 (1–3 ppm/yr, Buchwitz et al., 2007), although the observational period of the present study is more recent.

### **3.5** Comparison with $X_{CO_2}$ by FTS

 $X_{CO_2}$ (I) determined by the present method and  $X_{CO_2}$  by FTS (Bruker IFS 120 HR) were compared for the period from January 2007 to May 2008. Ohyama et al. (2009) retrieved CO<sub>2</sub> volume mixing ratios from solar absorption spectra in the 1.6-µm band measured with the ground-based high-resolution FTS at Tsukuba using profile retrieval and scaling retrieval algorithms. They derived the time series of  $X_{CO_2}$  from December 2001 to December 2007. In the present work, we extended the same analysis using the scaling retrieval algorithm to March 2008. Data on partly cloudy days were excluded by the same screening processes of Ohyama et al. (2009). Daily averaged  $X_{CO_2}$  by FTS are plotted in Fig. 2 and agree well with the present method in the autumn and winter seasons.

The peak-to-peak seasonal amplitude for  $X_{CO_2}$  using FTS measurements over Tsukuba between December 2001 and December 2007 was reported to be about 8 ppm by Ohyama et al. (2009). This peak-to-peak seasonal amplitude is larger than those from the present study reported in Sect. 3.4. Although the general features of the seasonal variations for the FTS measurements shown in Fig. 11 of Ohyama et al. (2009) and the present ones are similar, it would be necessary to extend the observational period before discussing differences in seasonal variations, as the observational period in the present study is much shorter than that in Ohyama et al. (2009).

#### 4 Conclusions

Column-averaged volume mixing ratios of  $CO_2(X_{CO_2})$  from January 2007 to May 2008 over Tsukuba were derived by using  $CO_2$  profiles measured by CONTRAIL.  $CO_2$  profiles from 493 flights on clear-sky days were analyzed. To calculate  $X_{CO_2}$ , two types of datasets, Tsukuba observational data (I) and global data (II), were alternatively used as ancillary data, and  $X_{CO_2}$ (II) with global data were compared with the Tsukuba observational data based on  $X_{CO_2}$ (I). The bias of  $X_{CO_2}$ (II) based on  $X_{CO_2}$ (I) over Tsukuba was derived to be -0.621 ppm with a standard deviation of 0.682 ppm. The uncertainty of  $X_{CO_2}$ (II) based on  $X_{CO_2}$ (I) was estimated to be 0.922 ppm, which is less than 0.3% of  $X_{CO_2}$ . The small uncertainty suggests that the present method of  $X_{CO_2}$  calculation using CONTRAIL data and the global data can be applied to airports worldwide. Therefore the number of validation sites of GOSAT for  $X_{CO_2}$  can be increased.

Acknowledgements. The authors wish to thank Takakiyo Nakazawa in Tohoku University for his helpful comment on the analysis. They are also grateful to anonymous reviewers and the editor for their careful reading of the manuscript.

Edited by: O. Wennberg

### References

- Aoki, S., Nakazawa, T., Machida, T., Sugawara, S., Morimoto, S., Hashida, G., Yamanouchi, T., Kawamura, K., and Honda, H.: Carbon dioxide variations in the stratosphere over Japan, Scandinavia and Antarctica, Tellus, 55B, 178–186, doi:10.1034/j.1600-0889.2003.00059.x, 2003.
- Bösch, H., Toon, G. C., Sen, B., Washenfelder, R. A., Wennberg, P. O., Buchwitz, M., de Beek, R., Burrows, J. P., Crisp, D., Christi, M., Connor, B. J., Natraj, V., and Yung, Y. L.: Space-based near-infrared CO<sub>2</sub> measurements: testing the Orbiting Carbon Observatory retrieval algorithm and validation concept using SCIA-MACHY observations over Park Falls, J. Geophys. Res., 111, D23302, doi:10.1029/2006JD007080, 2006.
- Buchwitz, M., Schneising, O., Burrows, J. P., Bovensmann, H., Reuter, M., and Notholt, J.: First direct observation of the atmospheric CO<sub>2</sub> year-to-year increase from space, Atmos. Chem. Phys., 7, 4249–4256, doi:10.5194/acp-7-4249-2007, 2007.
- COSPAR (Committee on Space Research): The COSPAR International Reference Atmosphere (CIRA-86), British Atmospheric Data Centre, http://badc.nerc.ac.uk/data/cira/, 2009.
- Dils, B., De Mazière, M., Müller, J. F., Blumenstock, T., Buchwitz, M., de Beek, R., Demoulin, P., Duchatelet, P., Fast, H., Frankenberg, C., Gloudemans, A., Griffith, D., Jones, N., Kerzenmacher, T., Kramer, I., Mahieu, E., Mellqvist, J., Mittermeier, R. L., Notholt, J., Rinsland, C. P., Schrijver, H., Smale, D., Strandberg, A., Straume, A. G., Stremme, W., Strong, K., Sussmann, R., Taylor, J., van den Broek, M., Velazco, V., Wagner, T., Warneke, T., Wiacek, A., and Wood, S.: Comparisons between SCIAMACHY and ground-based FTIR data for total columns of CO, CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O, Atmos. Chem. Phys., 6, 1953–1976, doi:10.5194/acp-6-1953-2006, 2006.
- Gurney, K. R., Law, R. M., Denning, A. S., Rayner, P. J., Baker, D., Bousquet, P., Bruhwiler, L., Chen, Y.-H., Ciais, P., Fan, S., Fung, I. Y., Gloor, M., Heimann, M., Higuchi, K., John, J., Maki, T., Maksyutov, S., Masarie, K., Peylin, P., Prather, M., Pak, B. C., Randerson, J., Sarmiento, J., Taguchi, S., Takahashi, T., and

Yuen, C.-W.: Towards robust regional estimates of  $CO_2$  sources and sinks using atmospheric transport models, Nature, 415, 626–630, doi:10.1038/415626a, 2002.

- Hamazaki, T., Kaneko, Y., Kuze, A., and Kondo, K.: Fourier transform spectrometer for greenhouse gases observing satellite (GOSAT), SPIE, 5659, 73–80, 2005.
- Inoue, H. Y. and Matsueda, H.: Variations in atmospheric CO<sub>2</sub> at the Meteorological Research Institute, Tsukuba, Japan, J. Atmos. Chem., 23, 137–161, 1996.
- Inoue, H. Y. and Matsueda, H.: Measurements of atmospheric CO<sub>2</sub> from a meteorological tower in Tsukuba, Japan, Tellus, 53B, doi:10.1034/j.1600-0889.2001.01163.x, 205–219, 2001.
- Intergovernmental Panel on Climate Change (IPCC), Climate Change 2007: The Physical Science Basis: contribution of Working Group I to the Fourth Assessment Report of the IPCC, Cambridge Univ. Press, New York, 2007.
- JMA: Japan Meteorological Agency, www.jma.go.jp/jma/indexe. html, 2009.
- Machida, T., Matsueda, H., Sawa, Y., Nakagawa, Y., Kondo, N., Goto, K., Nakazawa, T., Ishikawa, K., and Ogawa, T.: Worldwide measurements of atmospheric CO<sub>2</sub> and other trace gas species using commercial airlines, J. Atmos. Ocean. Tech., 10, 1744–1754, doi: 10.1175/2008JTECHA1082.1, 2008.
- Mao, J. and Kawa, S. R.: Sensitivity Studies for Space-Based Measurement of Atmospheric Total Column Carbon Dioxide by Reflected Sunlight, App. Optics, 43, 914–927, doi:10.1364/AO.43.000914, 2004.
- Matsueda, H., Machida, T., Sawa, Y., Nakagawa, Y., Hirotani, K., Ikeda, H., Kondo, N., and Goto, K.: Evaluation of atmospheric CO<sub>2</sub> measurements from new flask air sampling of JAL airliner observations, Pap. Meteorol. Geophys., 59, 1–17, doi:10.2467/mripapers.59.1, 2008.
- NCEP: NOAA/National Weather Service, National Centers for Environmental Prediction, NCEP Internet Services Team, www. ncep.noaa.gov/, 2009.
- NOAA (National Oceanic and Atmospheric Administration), NASA (National Aeronautics and Space Administration), US Air Force: US Standard Atmosphere, Washington, DC, 1976.
- Ohyama, H., Morino, I., Nagahama, T., Machida, T., Suto, H., Oguma, H., Sawa, Y., Matsueda, H., Sugimoto, N., Nakane, H., and Nakagawa, K.: Column-averaged volume mixing ratio of CO<sub>2</sub> measured with ground-based Fourier transform spectrometer at Tsukuba, J. Geophys. Res., 114, D18303, doi:10.1029/2008JD011465., 2009.
- Rayner, P. J. and O'Brien, D. M.: The utility of remotely sensed CO<sub>2</sub> concentration data in surface source inversions, Geophys. Res. Lett., 28, 175–178, 2001.
- Tateno Aerological Observatory, www.kousou-jma.go.jp/english/.
- Thoning, K. W., Tans, P. P., and Komhyr, W. D.: Atmospheric Carbon Dioxide at Mauna Loa Observatory 2, Analysis of the NOAA GMCC Data, J. Geophys. Res., 94, 8549–8565, 1989.
- Toon, G. C., Blavier, J.-F., Washenfelder, W., Wunch, D., Keppel-Aleks, G., Wennberg, P., Connor, B., Sherlock, V., Griffith, D., Deutscher, N., and Notholt, J.: Total Column Carbon Observing Network (TCCON), in Fourier Transform Spectroscopy, OSA Technical Digest (CD) (Optical Society of America, 2009), paper JMA3, http://www.opticsinfobase.org/abstract.cfm?URI= FTS-2009-JMA3, 2009 and Total carbon column observing network (TCCON), www.tccon.caltech.edu/, 2009.

- Washenfelder, R. A., Toon, G. C., Blavier, J.-F., Yang, Z., Allen, N. T., Wennberg, P. O., Vay, S. A., Matross, D. M., and Daube, B. C.: Carbon dioxide column abundances at the Wisconsin Tall Tower site, J. Geophys. Res., 111, D22305, doi:10.1029/2006JD007154, 2006.
- WMO: WMO Greenhouse Gas Bulletin, NO. 3, 2006, http://gaw.kishou.go.jp/wdcgg/products/bulletin/Bulletin2006/ ghg-bulletin-3.pdf, 2009.
- Yang, Z., Washenfelder, R. A., Keppel-Aleks, G., Krakauer, N. Y., Randerson, J. T., Tans, P. P., Sweeney, C., Wennberg, P. O.: New constraints on Northern Hemisphere growing season net flux, Geophys. Res. Lett., 34, L12807, doi:10.1029/2007GL029742, 2007.
- Yokota, T., Oguma, H., Morino, I., and Inoue, G.: A nadir looking SWIR FTS to Monitor CO<sub>2</sub> column density for Japanese GOSAT project, Proceedings of the Twenty-Fourth International Symposium on space Technology and Science, Miyazaki, Japan, 2004.
- Yokota, T., Yoshida, Y., Eguchi, N., Ota, Y., Tanaka, T., Watanabe, H., and Maksyutov, S.: Global Concentrations of CO<sub>2</sub> and CH<sub>4</sub> Retrieved from GOSAT: First Preliminary Results, SOLA, 5, 160–163, doi:10.2151/sola.2009 041, 2009.