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**CO₂ mixing ratio from
measurements by
commercial airlines**

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CO₂ column-averaged volume mixing ratio derived over Tsukuba from measurements by commercial airlines

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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 by using CO_2 concentration data observed by Japan Airlines Corporation (JAL) commercial airliners, based on the assumption that CO_2 profiles over Tsukuba and Narita were the same. CO_2 profile data for 493 flights on clear-sky days were analysed in order to calculate X_{CO_2} with an ancillary dataset: Tsukuba observational data (by rawinsonde and a meteorological tower) or global meteorological data (NCEP and CIRA-86). The amplitude of seasonal variation of X_{CO_2} (Tsukuba observational) from the Tsukuba observational data was determined by 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 and 381.7 ppm in May and September, respectively. The dependence of X_{CO_2} on the type of ancillary dataset was evaluated. The average difference between X_{CO_2} (global) from global climatological data and X_{CO_2} (Tsukuba observational), i.e., the bias of X_{CO_2} (global) based on X_{CO_2} (Tsukuba observational), was found to be -0.621 ppm with a standard deviation of 0.682 ppm. The uncertainty of X_{CO_2} (global) based on X_{CO_2} (Tsukuba observational) was estimated to be 0.922 ppm. This small uncertainty suggests that the present method of X_{CO_2} calculation using data from airliners and global climatological data can be applied to the validation of GOSAT products for X_{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 280–380 ppm (IPCC, 2007). This increase in CO_2 concentration enhances radiative forcing of the atmosphere and, thus, may

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contribute to climate change. The prediction of future atmospheric CO₂ concentration and its influence on climate will require accurate quantification of the distribution and variability of CO₂ sources and sinks, which have been derived from atmospheric CO₂ concentration data by using the inversion of atmospheric transport. Atmospheric CO₂ concentrations are measured with high accuracy mainly from ground stations. However, because of the sparseness of existing ground stations and the limitation of their altitudinal range, present estimates of CO₂ sources and sinks have large uncertainties (Gurney et al., 2002).

The usefulness of space-based observation for the source and sink estimates obtained by inversion has been evaluated. Rayner and O'Brien (2001) demonstrated that global space-based observation of monthly mean column-averaged CO₂ volume mixing ratios (VMR; precision $\leq 1\%$), described as X_{CO_2} , can be useful for reducing the uncertainties in regional ($8^\circ \times 10^\circ$ footprint) CO₂ source and sink estimates. Global X_{CO_2} values can be derived from space-based nadir-looking observation of sunlight scattered from the earth's surface in the near-infrared region (e.g., Mao and Kawa, 2004).

The Greenhouse gases Observing SATellite "IBUKI" (GOSAT) is used for space-based observation of the concentrations of CO₂ and methane (CH₄), two major greenhouse gases (Yokota et al., 2004, 2009; Hamazaki et al., 2005). By using the Fourier Transform Spectrometer (FTS) of the Thermal and Near-infrared Sensor for Carbon Observation (TANSO), which is an observational instrument onboard the satellite, X_{CO_2} and X_{CH_4} can be obtained as the globally observed products of the satellite.

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 were carried out between Scanning Imaging Absorption Spectrometer Atmospheric Chartography (SCIAMACHY) and ground-based FTS data obtained from 11 sites for total columns of CO, CH₄, CO₂, and N₂O (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

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Orbiting Carbon Observatory (OCO) retrieval algorithm and were compared with those obtained with SCIAMACHY (Bösch et al., 2006). The ground-based FTS is a powerful tool for the validation of satellite products. A network of ground-based FTSs that record direct solar spectra in the near-infrared spectral region, named TCCON (Total Carbon Column Observing Network, TCCON, Toon et al., 2009), provides essential validation data for SCIAMACHY and GOSAT. The network consists of 11 sites in Europe, Oceania, North America and Japan as of January 2009. 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 CO₂ profiles over 43 airports in the world from Narita International Airport (hereafter Narita) in Japan using five Japan Airlines (JAL) commercial airliners. Vertical CO₂ profiles are obtained during ascents and descents of the airliners and are useful to calculate X_{CO_2} over these airports. X_{CO_2} Values derived from the CONTRAIL data can be used to validate the X_{CO_2} data observed by GOSAT. The number of validation sites observed by the CONTRAIL project can be dramatically increased. To develop a method of X_{CO_2} calculation using CONTRAIL data, X_{CO_2} over Tsukuba calculated with CONTRAIL data over Narita should be evaluated as a first step, since there is a wealth of meteorological data for Tsukuba.

In the present work, first values of X_{CO_2} over Tsukuba during the period from January 2007 to May 2008 were derived, for which Tsukuba observational data were used as ancillary meteorological data. To calculate X_{CO_2} over other airports around the world, global meteorological data must be used in addition because there are limited numbers of nearby observational data. Thus, two types of datasets were alternatively used as ancillary meteorological data to calculate X_{CO_2} : Tsukuba observational data (from rawinsonde and a meteorological tower) and global meteorological data (National Centers for Environmental Prediction, NCEP¹, and Committee on Space Research, COSPAR,

¹NCEP: NOAA/National Weather Service, National Centers for Environmental Prediction, NCEP Internet Services Team, www.ncep.noaa.gov/

International Reference Atmosphere, CIRA,-86). The dependence of X_{CO_2} on the type of the dataset was derived, and the bias and uncertainty of X_{CO_2} (global) based on X_{CO_2} (Tsukuba observational) were estimated. Seasonal variation parameters of X_{CO_2} were obtained by a least-squares fitting to roughly evaluate the seasonal variation over
5 Tsukuba.

2 Analysis

In the present work, data from the Continuous CO₂ Measuring Equipment (CME) in CONTRAIL, which have an overall precision of 0.2 ppm (Machida et al., 2008), were mainly used to calculate X_{CO_2} . The data observed during the ascent and descent of
10 the airliners were taken as vertical CO₂ profiles over Narita. Two types of analyses using either Tsukuba observational data (type I analysis) or global meteorological data (type II analysis) as the ancillary meteorological data were performed to calculate X_{CO_2} over Tsukuba (Table 1).

To derive X_{CO_2} over Tsukuba, it was assumed that the CO₂ profile over Tsukuba
15 (36.1° N, 140.1° E) is the same as that over Narita (35.8° N, 140.4° E). Additionally, the CO₂ profiles in higher and lower altitude ranges from the observing altitude range of an airliner (from about 0.5–2 km to about 10–11 km) were assumed, as mentioned below.

Measurements showing that the stratospheric CO₂ concentration is constant above
20 an altitude of about 20 km are five years older (Aoki et al., 2003) than those of the global mean CO₂ concentration in the free troposphere, which was 381.2 ppm in 2006 with a growth rate of 1.9 ppm/yr (WMO, 2006), for example, 373.6 and 375.5 ppm in 2007 and 2008, respectively. When an airliner does not fly above the tropopause, the CO₂ concentration at the highest observational point (altitude) of an airliner was assumed to be constant up to the tropopause and the profile from the tropopause to 20 km is
25 linear 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 as shown by the dotted line in Fig. 1a. For a type I analysis, the height of the tropopause

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was obtained from a rawinsonde observation at the Tateno Aerological Observatory² (36.1° N, 140.1° E) of the Japan Meteorological Agency (JMA³), for which the lowest tropopause was selected from several observed tropopauses. Since the tropopause observations by rawinsondes were made twice a day at 00h00 and 12h00 UTC, the tropopause height at a given time was determined by linear interpolation. For a type II analysis, the tropopause height was obtained from the Model Analyses and Forecasts Global Forecast System (GFS) of the NCEP.

Under a planetary boundary layer (PBL), the CO₂ concentrations observed at a meteorological tower (36.1° N, 140.1° E, Inoue and Matsueda, 1996, 2001) in the Meteorological Research Institute (MRI), Tsukuba, Japan were used in the case of a type I analysis. The CO₂ 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 (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 CO₂ concentration at the lowest observational point (altitude 0.5~2 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 the NCEP, as systematic determination of the PBL height from rawinsonde data is difficult. The PBL height data from the NCEP taken every 3 h, at 00h00, 06h00, 12h00, and 18h00 UTC, with a 1° × 1° grid, were linearly interpolated with respect to the geographical coordinates of Narita and the takeoff or landing time. At 00h00, 06h00, 12h00 and 18h00 UTC analysed data in 0 h (the current time) of NCEP were used,

²www.kousou-jma.go.jp/english/index.HTM

³www.jma.go.jp/jma/indexe.html

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and at 03h00, 09h00, 15h00, and 21h00 UTC predicted data in three hours later. 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 air in type I analyses. The rawinsonde observations were made twice per day at 00h00 and 12h00 UTC. Rawinsonde data at 00h00 UTC were applied to airliner data collected between 18h00 UTC on the previous day and 06h00 UTC on the current day, whereas data at 12h00 UTC were applied to 06h00–18h00 UTC on the current day. Because the vertical resolution of rawinsonde observations is a few hundred metres, the number density at a specified altitude was obtained by logarithmic interpolation. Since rawinsonde data exist up to about 30 km, for higher altitudes over 30 km US standard atmosphere (NOAA, NASA, US Air Force, 1976) was utilized as the number density profile. In type II analyses, CIRA-86 data were employed. CIRA-86 data at latitude of every 5° were linearly interpolated 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 metres. The CO₂ concentration at an altitude of $100 \cdot n + 50$ m ($n=1, 2, 3, \dots$) was linearly interpolated with the two neighbouring observational points and the result was utilized as the concentration of the layer from $100 \cdot n$ to $100 \cdot (n+1)$ metres. As mentioned, with 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 0.5~2 km. In the case of the meteorological tower data, the CO₂ concentrations of the lowest three layers were estimated as follows:

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

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

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

where C_{layer} is the CO_2 concentration in the layer and C_{tower} is that by the tower. Numbers in parentheses for C_{layer} and C_{tower} indicate the altitudinal regions (in m) of the layers and observational heights (in m) in the tower, respectively. The coefficient of each term in Eqs. (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.

If the height of the lowest observational point of an airliner was higher than 4 km, the data of the flight were rejected from the analysis because of the data missing from the large altitudinal range from the PBL height to the lowest observational point. If a height of the highest observational point of an airliner was lower than 5 km, the data of the flight were also rejected because of the data missing in the higher region of the troposphere.

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 five airliners in clear-sky were analysed, for which the clear-sky condition was defined by an observation of the solar absorption spectra with an FTS in Tsukuba (Ohyama et al., 2009). X_{CO_2} from the ground level to an altitude of 85 km, i.e., the whole altitudinal range, and X'_{CO_2} for the altitudinal range of 2–10 km (hereafter X'_{CO_2}) were calculated as shown in Fig. 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

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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 differences between X_{CO_2} by CIRA-86 and by rawinsonde were derived for January 2007–May 2008 (Table 2). The average difference between $X_{\text{CO}_2}(I')$ and $X_{\text{CO}_2}(I)$, which is the bias of $X_{\text{CO}_2}(I')$ based on $X_{\text{CO}_2}(I)$, was found to be -0.043 ppm with a standard deviation of 0.067 ppm. The uncertainty of $X_{\text{CO}_2}(I')$ based on $X_{\text{CO}_2}(I)$ 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} .

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 around the PBL, i.e., that there is less influence 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 $X_{\text{CO}_2}(I)$ and $X_{\text{CO}_2}(II)$ were derived. The average difference, which is the bias of $X_{\text{CO}_2}(II)$ against $X_{\text{CO}_2}(I)$, was found to be -0.621 ppm with a standard deviation of 0.682 ppm. The uncertainty of $X_{\text{CO}_2}(II)$ based on $X_{\text{CO}_2}(I)$ was 0.922 ppm. X'_{CO_2} shows remarkably reduced uncertainties (Table 2), which may indicate small effects caused by CO_2 -profile turbulence around the PBL and small influences by air parcels that include high-concentration CO_2 from the Tokyo metropolitan area and a local CO_2 source. Furthermore, $X_{\text{CO}_2}(I')$ were compared with $X_{\text{CO}_2}(II)$ in order to subtract the dependence on air particle number-density datasets from the uncertainty of $X_{\text{CO}_2}(II)$ based on $X_{\text{CO}_2}(I)$. The differences between $X_{\text{CO}_2}(I')$ and $X_{\text{CO}_2}(II)$ were derived. The average of the

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differences, which is the bias of $X_{\text{CO}_2}(\text{II})$ against $X_{\text{CO}_2}(\text{I}')$, was found to be -0.578 ppm with a standard deviation of 0.691 ppm. The uncertainty of $X_{\text{CO}_2}(\text{II})$ based on $X_{\text{CO}_2}(\text{I}')$ was 0.901 ppm. Although the dependence on the number-density datasets was subtracted, the decrease in the uncertainty of $X_{\text{CO}_2}(\text{II})$ was small. Thus, the uncertainty of $X_{\text{CO}_2}(\text{II})$ 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 CO₂ source that disturbs the CO₂ profile uniformity around the PBL. In an analysis at another airport that has a uniform CO₂ profile around the PBL, the uncertainty of X_{CO_2} will be better than that of over Tsukuba.

3.3 Variability of X_{CO_2} within six hours around 13:00 LT

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 flight set during a window between 10h00 and 16h00 LT on a day in which the window was for 6 h at the center of the GOSAT overpass time (around 13h00 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_{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 km in X'_{CO_2}) did not yield a noticeable decrease in variability compared with the full altitudinal range (X_{CO_2}). The variabilities depend on the time windows originated mainly from the variability of the CO₂ profiles observed by airliners.

3.4 Screening criteria

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.

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Flights from/to Narita can be classified into the following two cases: airliners that take off and land in the northern (case N) and southern (case S) airspaces of the airport. The observational points below an altitude of 1 km for cases N and S were 20–30 and 50–60 km from Tsukuba, respectively. The high concentrations of X_{CO_2} of more than 382.5 ppm that were recorded in August 2007 were almost always from those of case S. CO₂ profiles with a high concentration of X_{CO_2} show high concentrations of CO₂ in the low altitudinal region (0–2 km). These may be classified into the following three cases: high concentrations of CO₂ observed by aircrafts (case 1), in the MRI tower (case 2), or in both (case 3). If the condition of a flight is in case S and case 1, air parcels having high concentration of CO₂ over the Tokyo metropolitan area are not inflowing over Tsukuba and the assumption of uniformity for the CO₂ profile over Tsukuba and Narita cannot be approved. Flight data for the condition, i.e. case S and case 1, should not be considered for the validation of GOSAT data. In the present work, only one flight on 16 August 2007 that had X_{CO_2} of 388.5 ppm was in the condition to show the CO₂ profile in Fig. 3.

3.5 Amplitude of seasonal variation

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

$$f(t) = a_1 + a_2 \cdot t + a_3 \cdot t^2 + a_4 \cdot \sin(2\pi t) + a_5 \cdot \cos(2\pi t) + a_6 \cdot \sin(4\pi t) + a_7 \cdot \cos(4\pi t), \quad (4)$$

where t is a variable of time in the unit of years from the starting date of the computation on 1 January 2003. Since the obtained X_{CO_2} data in the present work were limited to one year and three months, the coefficient a_3 was fixed to 0.0. The values of a_1 , a_2 , a_4 , a_5 , a_6 , a_7 were determined by a least-squares fit. The datum of the flight of case S and case 1, as mentioned in Sect. 3.4, was not included in the fit. The standard deviation of the differences between X_{CO_2} and the fitting curve was 0.98 ppm (0.74 ppm for X'_{CO_2}). The obtained coefficients are listed in Table 3 and the fitting curves of X_{CO_2} and X'_{CO_2} are shown in Fig. 2. The highest and lowest values of the fitting curve

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in 2007 were 386.4 and 381.7 ppm for X_{CO_2} and 387.0 and 381.1 ppm for X'_{CO_2} in May and September, respectively. The amplitude of the tentative seasonal variation, i.e., the peak-to-peak seasonal amplitude, by the present observation period for one year and three months was found to be 4.63 and 5.91 ppm by the fitting curves for X_{CO_2} and X'_{CO_2} , respectively. The peak-to-peak seasonal amplitude for X_{CO_2} using FTS (Bruker IFS 120 HR) measurements over Tsukuba between December 2001 and December 2007 was reported to be about 8 ppm by Ohyama et al. (2009). 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).

The values determined for a_2 (2.27 and 2.45 ppm/yr) show tentative growth rates for X_{CO_2} and X'_{CO_2} , respectively, 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 observation at northern 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 than those of these other two sets of measurements.

4 Conclusions

Column-averaged volume mixing ratios of CO₂ (X_{CO_2}) from January 2007 to May 2008 over Tsukuba were derived by using CO₂ concentration data observed by CONTRAIL. CO₂ profile data from 493 flights on clear-sky days were analysed. To calculate X_{CO_2} , two types of datasets, Tsukuba observational data (I) and global data (II) as ancillary data, were alternatively used, and X_{CO_2} (II) with global data were compared with the

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Tsukuba observational data based on $X_{\text{CO}_2}(\text{I})$. The bias of $X_{\text{CO}_2}(\text{II})$ based on $X_{\text{CO}_2}(\text{I})$ over Tsukuba was derived to be -0.621 ppm with a standard deviation of 0.682 ppm. The uncertainty of $X_{\text{CO}_2}(\text{II})$ based on $X_{\text{CO}_2}(\text{I})$ 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 subservient global data can be applied to airports worldwide. Therefore, the number of validation sites of GOSAT for X_{CO_2} can be increased.

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Table 1. Type of analysis of X_{CO_2} for ancillary meteorological and model data.

	Analysis Type		
	I	I'	II
MRI CO ₂	Yes	Yes	No
Number density profile	Sonde	CIRA-86	CIRA-86
NCEP PBL	Yes	Yes	No
Tropopause	Sonde	Sonde	NCEP

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Table 2. Uncertainties of X_{CO_2} based on type I analysis (ppm). To show small relative values, the uncertainties are given to three decimal places.

	Altitudinal range		Analysis Type			
			I	I'	II	II-I'
X_{CO_2}	whole	Bias	0.0	-0.043	-0.621	-0.578
		Error	0.0	0.067	0.682	0.691
		Uncertainty	0.0	0.080	0.922	0.901
X'_{CO_2}	2–10 km	Bias	0.0	-0.018	-0.019	-0.001
		Error	0.0	0.029	0.037	0.022
		Uncertainty	0.0	0.034	0.041	0.022

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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 ^a	Unit	Whole ^b	2–10 km
a_1	ppm	374.1929	373.1888
a_2	ppm/yr	2.2672	2.4515
a_3	ppm/yr ²	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
a_7	ppm	0.2523	−0.0222

^a These are the coefficients in Eq. (4). The coefficient a_3 was fixed to 0.0. See text for details.

^b Whole altitudinal range, i.e., from the ground level to the lower thermosphere (85 km).

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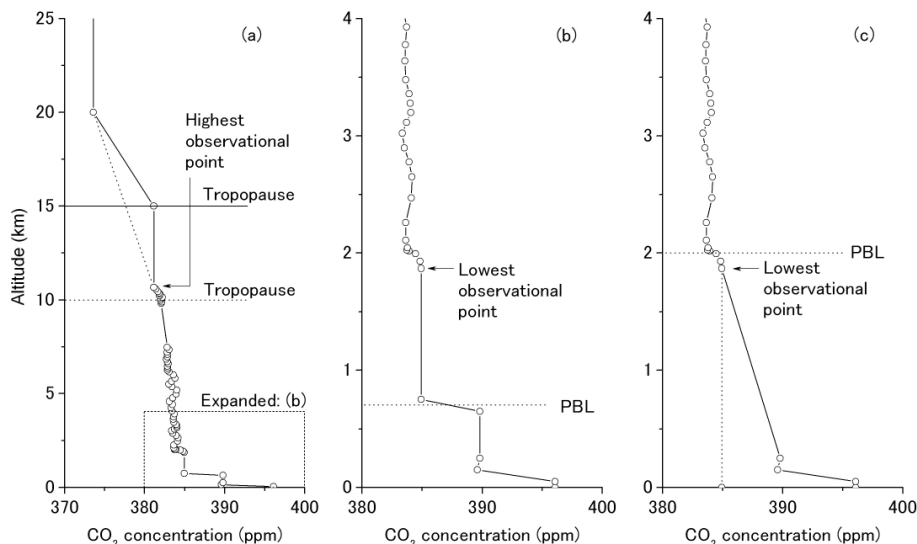


Fig. 1. CO₂-profile assumption over Tsukuba. **(a)** High-altitude profiles: the solid and dotted lines show cases in which the tropopause is higher and lower, respectively, than the highest observational point by an airliner. The rectangular area is expanded in **(b)** and **(c)**. **(b)** Low-altitude 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.

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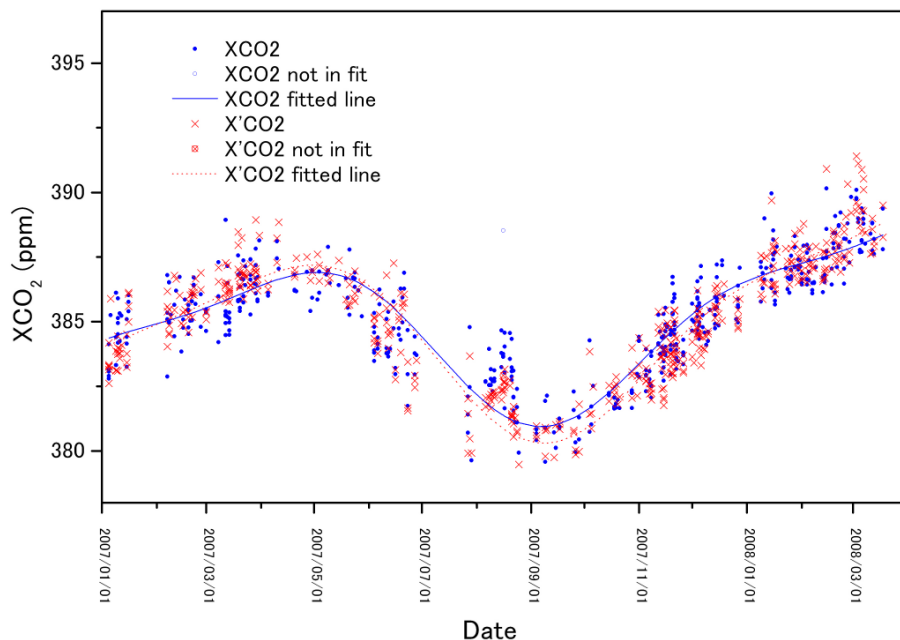


Fig. 2. Time series of X_{CO_2} and X'_{CO_2} from type I analysis over Narita using CONTRAIL data from January 2007 to May 2008. Data from 493 flights by five airliners were analysed. X_{CO_2} (blue marks and a blue line) were numerically integrated to cover the whole altitudinal range, i.e., from the ground level to the lower thermosphere (85 km), and for X'_{CO_2} (red marks and a red line) over the altitudinal range of 2–10 km. Data on 16 August 2007 were not in fit.

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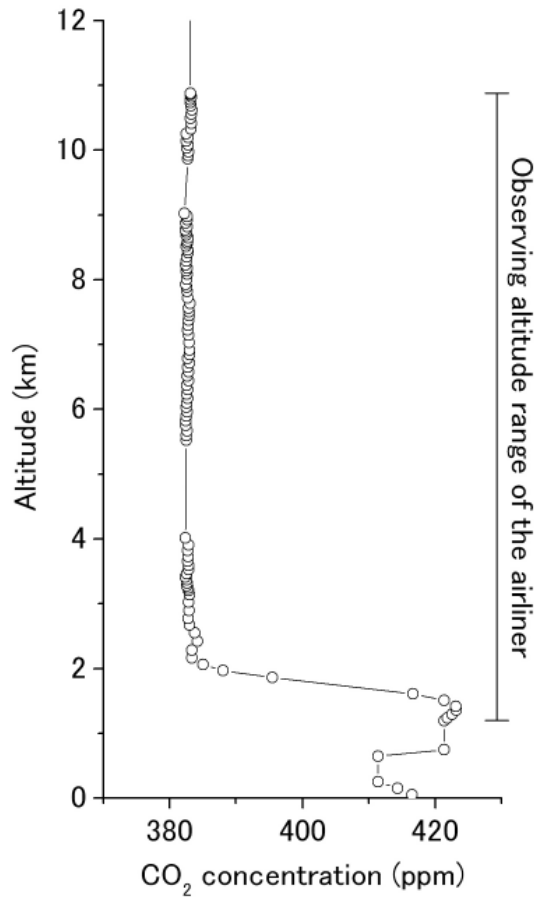


Fig. 3. An example of a profile of an inflowing air parcel from over the Tokyo metropolitan area to over Narita on 16 August 2007.

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