

Relative contribution of transport/surface flux to the seasonal vertical synoptic CO₂ variability in the troposphere over Narita

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ABSTRACT

Frequent CO₂ measurements obtained by commercial aircraft provide a unique, quasi-continuous record of free-tropospheric CO₂ variability. Vertically-resolved synoptic-scale fluctuations of CO₂ over Narita International Airport (lat 35.8°N, 140.4°E, 43 m above sea-level) were investigated from November 2005 to March 2009, and combined with analyses of results from a transport model simulation for the year 2007 to retrieve information on sources contributing to the observed variability. The synoptic-scale variability of the observed CO₂ mixing ratio, represented by the standard deviation (SD) from the fitted curves, increased in the upper troposphere in the spring, with a noticeable increase at all altitudes in the summer. This seasonal/altitudinal change of the observed SD was shown to be statistically significant throughout the observation period, and the model result agreed with the observation except for the underestimation of the summertime SD. Tagged simulations were conducted to evaluate the relative contribution of the regional fluxes to the synoptic-scale variability over Narita. The results indicate that the major contribution to the free troposphere (FT) variability was made by the fluxes in East Asia, while the Japanese fluxes contributed mostly to the variability in the planetary boundary layer (PBL). A sensitivity analysis was performed to evaluate the relative influence of transport and of flux magnitude on the CO₂ SD over Narita for 2007. It was found that a change in the surface flux magnitude could affect the altitudinal distribution of the annual SD over Narita as follows: 41 and 3% at 9 km, 61 and 4% at 5 km, 19 and 83% at 0.5 km when the fossil fuel flux from East Asia and Japan was doubled, respectively. These results are qualitative in nature (since SD is a non-linear function of concentration and flux), but do indicate that the CO₂ SD over Narita is more sensitive to the fluctuation in the atmospheric transport (synoptic-scale meteorological variability) in the FT, while showing much more sensitivity to the magnitude of local fluxes in the PBL. The results also point to the fact that vertical profiles of atmospheric CO₂ variability at the synoptic scale could potentially provide a useful additional constraint in the inversion analysis of regional CO₂ fluxes.

Keywords: synoptic-scale variability, carbon dioxide, vertical profile, transport model, free troposphere, regional fluxes, aircraft measurements

1. Introduction

Atmospheric concentration of CO₂ shows variation over a range of scales, from inter-annual and seasonal to synoptic and diurnal. Inter-annual changes result mainly from fluctuations in the global flux balance, while the seasonal

cycle reflects the seasonal variation in the flux distribution of sources and sinks. The short time-scale variability of atmospheric CO₂ can behave as a ‘noise’ against the signal produced by the surface flux and has to be detected from the monthly averages of the total CO₂ concentration (Karstens et al., 2006). However, the synoptic-scale variability can be a good indicator of regional surface CO₂ fluxes, of the strength of vertical mixing, and of atmospheric transport by synoptic meteorological systems.

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The synoptic scale variability of the CO₂ concentration has been studied mostly using boundary-layer data (Chan et al., 2004; Law et al., 2008; Parazoo et al., 2008; Patra et al., 2008). While the seasonal and latitudinal variation of CO₂ above the boundary layer have been systematically investigated by periodic airborne sampling programmes (Tanaka et al., 1983; Tanaka et al., 1988; Nakazawa et al., 1991; Nakazawa et al., 1993; Anderson et al., 1996; Matsueda and Inoue, 1996; Matsueda et al., 2002; Vay et al., 2003; Geels et al., 2007), studies on various aspects of the synoptic-scale variability have been sparse due to the lack of periodic high-frequency measurements. Although there have been several intensive aircraft sampling programmes recently (Lin et al., 2006; Shashkov et al., 2007; Choi et al., 2008), those are still mostly restricted to individual campaigns employing research aircrafts. In this study, we explore the synoptic-scale variability observed in the vertical CO₂ profiles obtained frequently by commercial aircraft. The observations have been made regularly during all seasons, and show a clear indication of a seasonal variation in the synoptic-scale variability. We compared observations with results from global atmospheric transport model simulations forced by prescribed CO₂ flux distributions (fossil fuel (FF), terrestrial biosphere (TB), and ocean). After validating the ability of the model to realistically reproduce synoptic-scale variability of CO₂ in the troposphere, we proceed to discuss the controlling factors of the variability in tropospheric CO₂ over Narita, by reference to the result of a tagged model simulation that distinguishes CO₂ fluxes from different regions. The results of our study contribute further insight into the range of uncertainty associated with constraining regional-scale fluxes of CO₂ with seasonal variation in the vertical CO₂ distribution. The results of the study also provide some useful information on the retrieval of atmospheric CO₂ concentrations using satellite spectral data (Eguchi et al., 2010; Yoshida et al., 2011).

2. Experiment

The observational data used in this study were derived from frequent measurements of CO₂ over Narita Airport from November 2005 to March 2009, obtained as a part of the project called Comprehensive Observation Network for TRace gases by AIrLiner (CONTRAIL) (Machida et al., 2007; Machida et al., 2008; Matsueda et al., 2008; Sawa et al., 2012). Onboard measurements of CO₂ were made by the Continuous CO₂ Measuring Equipment (CME) using a non-dispersive infrared gas analyser (NDIR). CME is calibrated periodically by using two standard gases and its accuracy is kept within 0.2 ppm. Since our focus is on the synoptic-scale variability of CO₂ and its vertical profiles, we chose for our study the Narita International Airport

(NRT), over which we have the most frequent measurements compared to other locations. NRT is the busiest air-freight hub in Japan, located approximately 60 km east of downtown Tokyo (Fig. 1). Ten-second averaged data are recorded on the aircraft during the ascent from and descent into Narita. A total of 1044 vertical profile observations were made over Narita in 2007; two to three profiles per day on average. There were nine data gaps longer than 2 d throughout the year, with the longest gap occurring for 10 d from 3–12 October.

Tagged tracer simulations were conducted using the National Institute for Environmental Studies global transport model (NIES TM) version 05 with a horizontal resolution of $1^\circ \times 1^\circ$ and vertical resolution of 47σ levels from the surface to 0.01σ (Maksyutov et al., 2008). This is an off-line model driven by meteorological data from the National Center for Environmental Prediction (NCEP) Global Forecast System (GFS) at 3-h time step and $1^\circ \times 1^\circ$ horizontal resolution. In our study, the input CO₂ flux data (FF, TB, and ocean) were taken from the protocol for the CONTRAIL transport model intercomparison experiment (Niwa et al., 2011). The FF emission is derived from the EDGAR-1998 (Olivier and Berndowski, 2001) distribution and the country-specific FF consumptions from CDIAC (Boden et al., 2009). For the TB, we generated 3-hourly fluxes from monthly mean fluxes of the Carnegie-Ames-Stanford Approach (CASA) model (Randerson et al., 1997) using a procedure similar to that of Olsen and Randerson (2004) with meteorological fields from the Japan Meteorological Agency Climate Data Assimilation System (JCDAS) (Onogi et al., 2007). The ocean flux was taken from Takahashi et al. (2009). No inverse fluxes were used in this study.

Tags were put on tracers from six regions (Japan, East Asia, East Russia, South-east Asia, India, and Himalaya) as shown in Fig. 1. We considered these regions to constitute a substantive portion of the CO₂ footprint for Narita, basing our assumption on the idea that the flux signals from regions farther away would be smoothed out by atmospheric turbulence and mixing during the long-range transport, washing out any distinctive flux signature. For each simulation, we used the last year of a 5-yr model run driven by the NCEP meteorological field for 2007. The model output interval of the CO₂ concentration field was 4 h. The CO₂ values of the model output are given as deviations from an arbitrary initial value, and not as absolute concentration values. This did not impact our analysis since the focus of our study was on variability.

Observed and calculated CO₂ mixing ratios over the grid box that encompassed NRT were divided into 2-km thick altitudinal bins (0–2, 2–4, 4–6, 6–8, 8–10 km). Measurements that were taken in the lower troposphere were judged to be inside the planetary boundary layer (PBL) if the

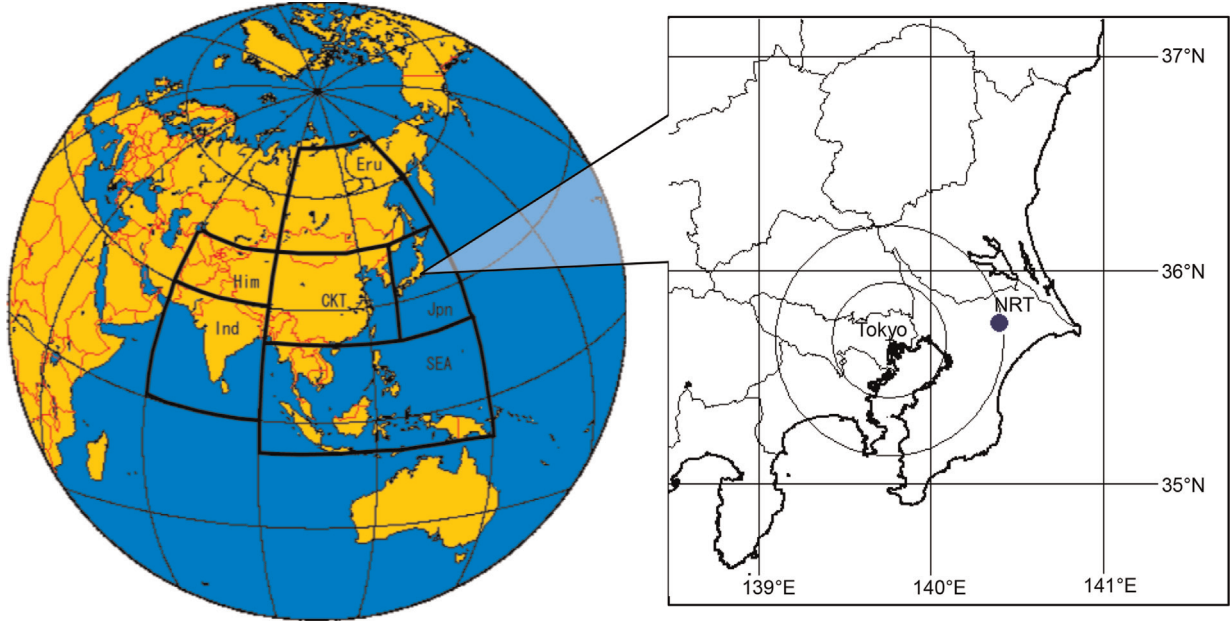


Fig. 1. Geographic setting of the simulations. Left: boundaries and names of the six tagged regions; Japan (Jpn), East Asia (CKT), East Russia (Eru), South-east Asia (SEA), India (Ind), and Himalaya (Him). Right: Enlarged view of the Kanto plain, showing the location of the Narita International Airport (NRT). The inner and outer concentric circles indicate distances of 30 and 60 km from downtown Tokyo, respectively.

altitudes of the measurements were below the height of the mixed layer estimated every 6 h. The mixed layer is defined as having a bulk Richardson number value of less than 0.25 (Troen and Mahrt, 1986). According to the method used by Sawa et al. (2012), the CO₂ data in the layer with a difference of potential temperature of up to 10 K with the top of the PBL were not used in this study. The potential vorticity analysis method used by Sawa et al. (2008) was applied to determine those data that were located in the stratosphere. For the data in each altitudinal bin, a daily mean standard deviation (SD), which was used as a metric for CO₂ variability, was calculated using annual and seasonal detrended data. To remove the trend and seasonal cycle, the digital filtering technique of Nakazawa et al. (1997) was applied, with cut-off frequencies of 24 months and 4 months for the long term trend and for the short term variation, respectively.

3. Results and discussions

3.1. Vertical seasonal variation of the synoptic CO₂ variability over Narita

Observed and simulated time series of CO₂ mixing ratios for 2007 at an altitude of 5 km over Narita are shown in Fig. 2. In addition to the distinct seasonal variation mainly due to the TB, we can see significantly large temporal variations on daily to weekly time-scales (grey line). The

day-to-day variation, shown as the monthly SD, is denoted by the error bars in Fig. 2. The length of the error bars varies considerably throughout the year, indicating seasonal dependence of the magnitude of the CO₂ concentration fluctuation at the synoptic time-scale. These fluctuations are influenced by the seasonal variations in the atmospheric transport and in the surface CO₂ flux. The importance of these two factors and their respective contribution to the observed vertical profiles of synoptic fluctuations in the CO₂ concentration at Narita is discussed below.

Figure 3 shows the seasonal differences in the vertical profiles of CO₂ SD obtained from the observation and the simulation. Each value corresponds to the mean SD of each of the 2-km altitude bins, calculated for four seasons (MAM: March-April-May, JJA: June-July-August, SON: September-October-November, DJF: December-January-February). Except for the bottom bin, the bin average is given at the mid-level of the bin thickness (3 km for the 2–4 km bin, 5 km for the 4–6 km bin, 7 km for the 6–8 km bin, and 9 km for the 8–10 km bin). As 97% of the observed data below 2 km were categorised as the PBL data, the SD for the lowest altitude bin (0–2 km) was derived from the PBL data only. The observations made at altitudes higher than 10 km or categorised as stratospheric data were eliminated when deriving the SD for the highest altitude bin (8–10 km). The SDs observed in the PBL were 3–7 ppm, which was much higher than the free troposphere (FT) value of 1–2 ppm. The SDs derived for each month

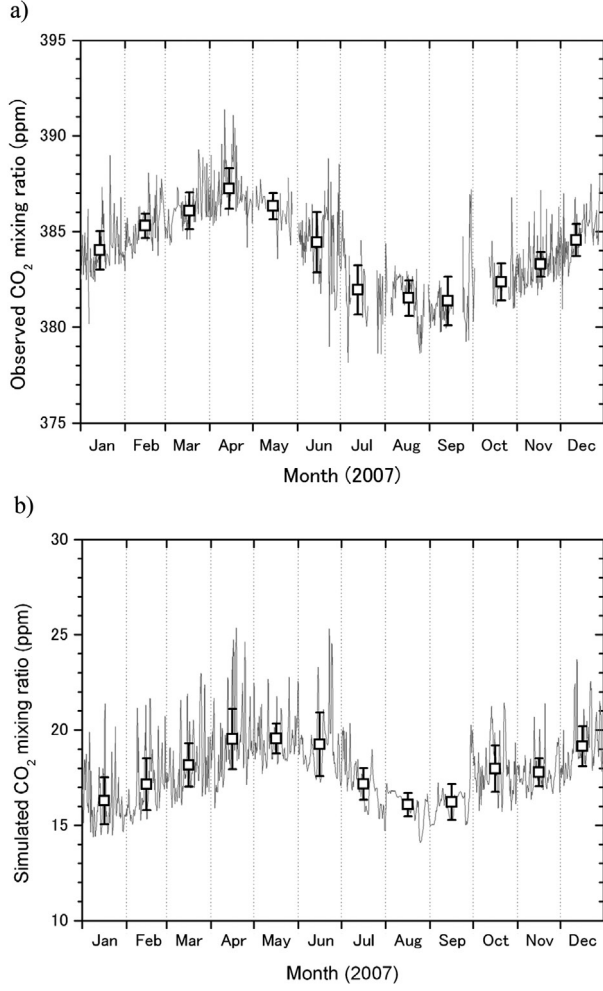


Fig. 2. Time series of CO_2 mixing ratios in 2007 observed (a) and simulated (b) at the 5-km altitude level over Narita. The line plot shows the daily variation, the open squares show the monthly mean, and the error bars show the monthly standard deviation. The model output is not given as absolute values of the CO_2 mixing ratio but as the temporal variation of the deviation from an arbitrary initial value.

from the observed and simulated CO_2 are shown in Tables 1 and 2, respectively. For the observation data, the SD was not calculated when the observation frequency (the number of observed days per total number of days in the month) was less than 30%. The dataset in the PBL in October 2007 did not meet this criterion, hence it is stated as not available (NA) in Table 1. Since the purpose of this study was to obtain an overview of the seasonal/vertical change of CO_2 synoptic variability, we used seasonally aggregated data for the analysis if not otherwise specified.

The vertical profiles of observed SDs did not show large seasonal variations except for the increase at all altitudes in the summer and in the high altitude bin in the spring (Fig. 3). This seasonal/altitudinal change in the observed

SD was statistically significant at the 99% confidence level throughout the observation period. The elevated SD can be explained by a combination of seasonal changes in the atmospheric dynamics and in the CO_2 fluxes. Although the model data agreed relatively well with the observed data in reproducing the SD over a 1-yr period, the observed SDs were underestimated by the model for the summer season. This discrepancy will be investigated in a later section.

In addition to the seasonal/altitudinal change of observed and simulated SDs, we also checked the correlation between the observed and simulated daily deviation time series for a period of 1 yr in five altitude bins. The correlation coefficients $R = 0.18, 0.54, 0.53, 0.56$ and 0.52 for the PBL and the 2–4 km, 4–6 km, 6–8 km and 8–10 km levels, respectively, were found to be statistically significant

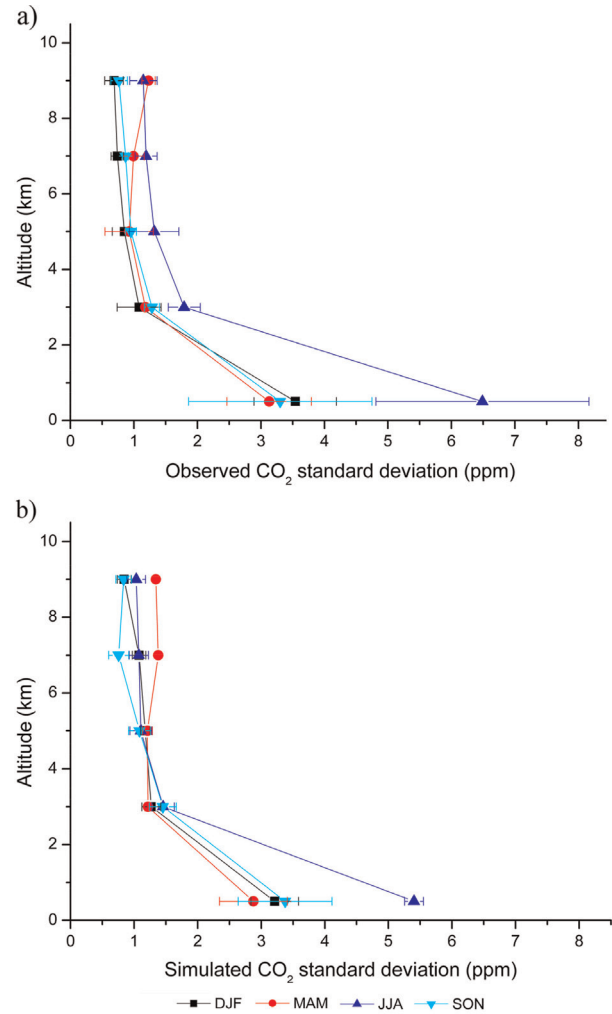


Fig. 3. Vertical profiles of seasonal mean standard deviation of CO_2 mixing ratios from a) CONTRAIL aircraft observations in 2007, b) simulations for the year 2007 using the NIES-TM version 05. The error bars indicate the inter-annual standard deviation for 2006–2008.

Table 1. Vertical/monthly mean standard deviation of CO₂ (ppm) observed over Narita

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
PBL	2.67	3.73	2.36	3.68	3.63	8.18	6.33	4.35	3.99	NA	3.16	4.28
5 km	1.01	0.64	0.96	1.06	0.68	1.58	1.28	0.93	1.26	0.96	0.64	0.84
9 km	0.73	0.64	1.45	1.24	0.94	1.26	1.06	1.09	1.03	0.62	0.61	0.65

The standard deviation was not calculated for October in the planetary boundary layer (PBL), following the criterion based on time coverage and number of observations (see Section 3.1).
NA = not available.

at the 90% confidence level. The seasonal correlation coefficients R in the PBL were 0.40, 0.28, 0.01 and 0.25 for winter, spring, summer and autumn, respectively. Since the predominant northern Pacific high pressure system creates a stagnant condition typical of summertime weather in Japan, Narita is occasionally affected by extremely high concentrations of CO₂ emitted from the Tokyo Metropolitan Area, depending on the orientation of the sea-land breeze circulation system. The topography of the area is very complicated, which leads to complex wind patterns. The lower correlation coefficient in the PBL indicates that the model could not always reproduce the fine spatial temporal variation, which is affected by nearby sinks/sources, local wind, topography and land use. Especially in the summer, a higher resolution is needed to improve the simulation of CO₂ in the PBL (Maksyutov et al., 2008).

3.2. Result of the tagged simulation experiment

3.2.1. Flux sources and regions governing the SD of CO₂ over Narita. Figure 4 shows the results from the tagged simulation experiment, showing seasonal vertical profiles of the CO₂ SD caused by different flux sources from different regions. For the purpose of our discussion, we divide the vertical SD distribution into two layers: the bottom layer (<2 km) corresponding to the PBL and the top layer (>2 km) corresponding to the FT.

In the bottom layer (<2 km), the variation of FF CO₂ from Japan contributed the most throughout the year. This indicates that the SD in the bottom layer is controlled mainly by the variability in the surface flux near the observation point and by the local meteorological conditions (wind direction, cyclone passage, etc.). The underestimation of the simulated SDs in the bottom layer points to insufficient resolution of subgrid variations in surface

fluxes and transport in the model. Since the temporal variation of the flux is also an important potential contributor to the simulated SD, it is very possible that the underestimation by the model was caused, at least in part, by the use of the time-invariant, constant FF fluxes throughout the year. Seasonal variations of the observed SD in the bottom layer showed considerably higher values during the summer season compared to other seasons.

In the top layer (>2 km), the observed SD stayed vertically invariant at about 1 ppm except for a ~ 0.7 ppm increase seen in the summer and in the spring (Fig. 3). While the enhancement was constant throughout the top layer in the summer, the springtime enhancement increased with altitude (Fig. 3). As shown in Fig. 4, the influence from sources in Japan diminished rapidly with altitude and almost disappeared above 4 km. Meanwhile, the fluxes from East Asia contributed the most throughout the year in the top layer. This result is consistent with the fact that Narita is located just downwind from one of the high-emission regions in the world.

3.2.2. Correlation between the biosphere-CO₂ and FF-CO₂ over Narita. Land vegetation goes through a seasonal cycle of photosynthetic absorption of atmospheric CO₂ while the FF flux remains relatively constant through the year. This causes a vertically and seasonally varying relationship between the TB-related CO₂ and the FF-related CO₂, modulated at all times by the vertical mixing time-scale of about a month. Here we did not focus on the sign of the deviation but only on the correlation in order to follow the vertical/seasonal propagation of the effect of surface fluxes up in the air. As shown in Table 3, the correlation between the TB-CO₂ and the FF-CO₂ over Narita showed a relatively high positive correlation

Table 2. Vertical/monthly mean standard deviation of CO₂ (ppm) simulated over Narita

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
0.5 km	3.18	2.89	2.25	2.95	3.11	5.08	6.44	4.38	3.81	2.74	3.72	3.57
5 km	1.24	1.36	1.14	1.58	0.79	1.68	0.83	0.62	0.94	1.22	0.73	1.05
9 km	0.74	1.00	1.26	1.48	1.32	1.63	0.68	0.36	0.63	0.64	1.20	0.92

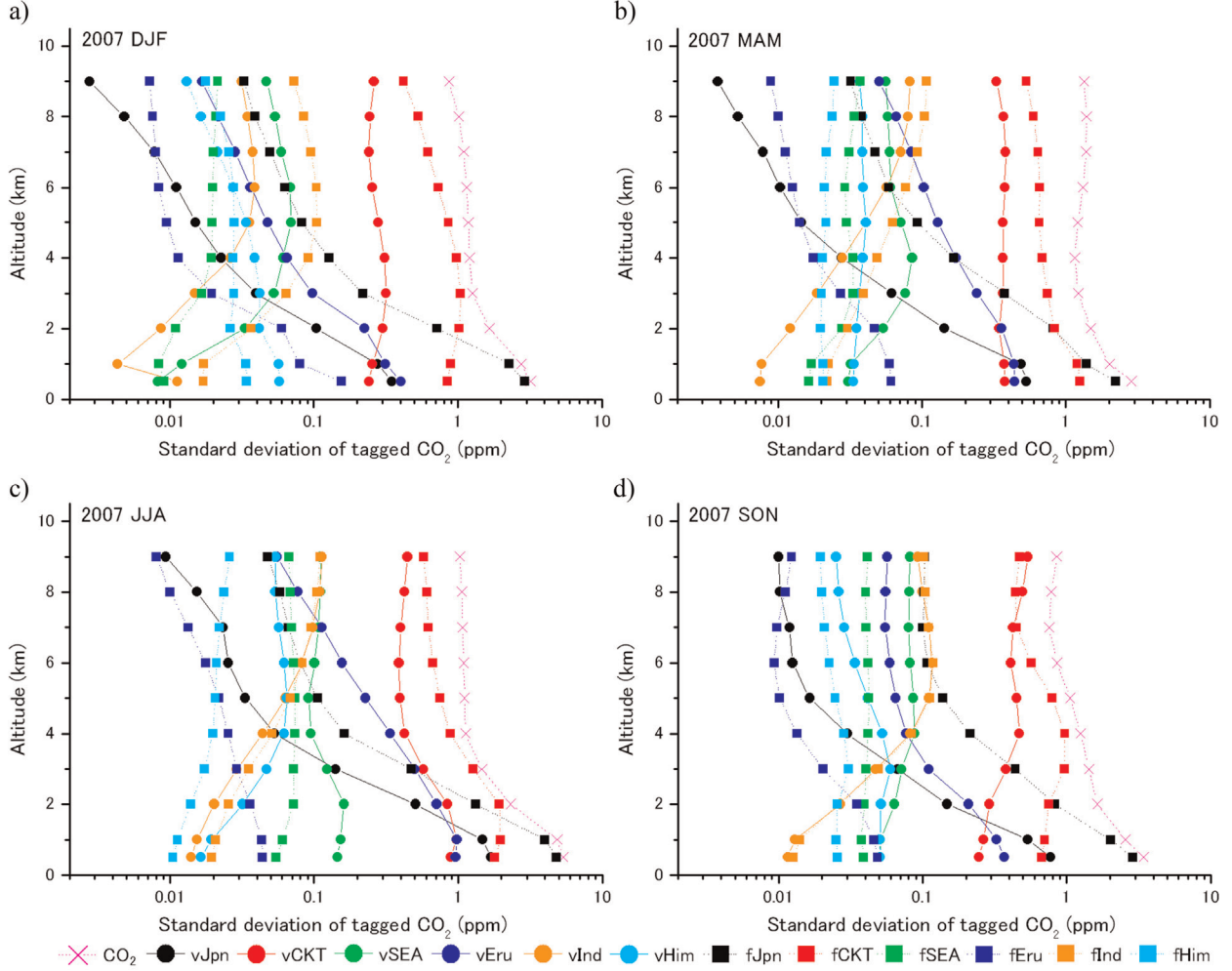


Fig. 4. Vertical profiles of the four seasonal mean standard deviations of the CO₂ mixing ratios in 2007 from the tagged simulation experiment using the NIES-TM version 05. The scale is logarithmic. The description of the tracer names is given in Table 4.

throughout the FT in the winter (0.69–0.81). With the arrival of spring, the correlation in the 2–4 km bin started to decrease as the TB in the Northern Hemisphere began to absorb CO₂, becoming negative in the summer. As the seasonally low CO₂ concentration in the PBL propagated upward, the correlation became progressively negative through the depth of the troposphere over Narita, with

Table 3. Vertical/seasonal correlation coefficients (R) of the standard deviation of CO₂ originating from the global fossil fuel (FF) flux and the global terrestrial biosphere (TB) flux in the free troposphere (FT) over Narita

	DJF	MAM	JJA	SON
3 km	0.81	0.64	−0.61	0.14
5 km	0.71	0.76	−0.37	0.15
7 km	0.72	0.78	−0.08	−0.12
9 km	0.69	0.77	−0.12	−0.35

the highest negative values observed in the lower troposphere (−0.61). As the season progressed into autumn, the negative correlation values propagated upward, reaching the largest negative value of −0.35 in the 8–10 km bin. In the meantime, the correlation in the lower troposphere became positive again, as photosynthesis became small and the respiratory CO₂ emission began to dominate. Table 3, along with the results from Figs. 3 and 4, indicates the seasonally-varying influence of transport from different CO₂ sources to the CO₂ variability observed over Narita. This relationship plays one of the key roles in the sensitivity study described in section 3.3.

3.2.3. Increase of SD in the upper troposphere in spring.

The results from the tagged simulation clearly showed enhanced transport of CO₂ from East Asia to the upper troposphere over Narita in the spring, suggesting an active

Table 4. Names of the tracers used in the tagged-tracer simulation

Tracer name	Flux type and region
fJpn	Fossil fuel from Japan
vJpn	Vegetation from Japan
fCKT	Fossil fuel from East Asia (China + Korea + Taiwan)
vCKT	Vegetation from East Asia (China + Korea + Taiwan)
fSEA	Fossil fuel from South-east Asia
vSEA	Vegetation from South-east Asia
fEru	Fossil fuel from East Russia
vEru	Vegetation from East Russia
fInd	Fossil fuel from India
vInd	Vegetation from India
fHim	Fossil fuel from Himalaya
vHim	Vegetation from Himalaya

Description of fluxes is given in Section 2. The geographical setting and the boundaries of the regions are shown in Fig. 1.

passage of Asian outflow (Fig. 5). There have been numerous observational and modelling studies about intercontinental-scale transport of atmospheric tracers in the FT (e.g. Jaffe et al., 1999). The strongest Asian outflow occurs in the spring due to an increased number of eastward-tracking synoptic systems that allow an enhanced ventilation of the continental boundary layer. This is reinforced by the efficient transport of stronger mid-latitude westerly winds in the FT as the receding Siberian high is gradually replaced by the northward moving subtropical high pressure system. The observed higher SD at higher altitudes in the spring also indicates the active passage of the Asian outflow in the middle and upper troposphere (Uno et al., 2009).

3.2.4. Increased CO₂ variability in summer. As shown earlier, the greatest CO₂ variability throughout the depth of the troposphere was observed in the summer season (defined as June-July-August). However, the increase in the observed SD occurs earlier in the FT than in the PBL. In the early summer, East Asia is under the summer monsoon called the Baiu in Japan (Sampe and Xie, 2010). In 2007, the Baiu started in mid-June and lasted until the end of July. Figure 2 indicates that the increase in SD in the FT (5-km level) coincided with the beginning of the Baiu. On the other hand, the increase in SD in the PBL did not begin until after the Baiu had finished. This can be explained by the fact that, in August, after the Baiu season, a stable high pressure system (subtropical Pacific high) settles in, periodically bringing air with high CO₂ concentration from the Tokyo district to Narita under the prevailing southerly to south-westerly winds (Araki et al.,

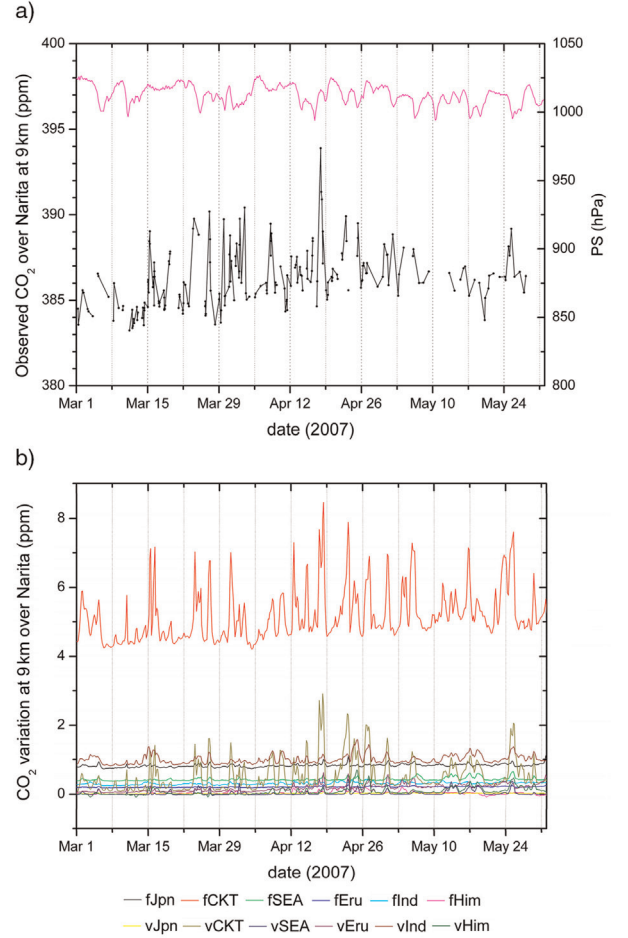


Fig. 5. Time series of a) observed CO₂ (scale on the left) and surface pressure (scale on the right) and b) simulated CO₂ from each tagged flux in the 8–10-km altitude range over Narita. The description of the tracer names is given in Table 4.

2010). Under stagnant conditions, negative deviations were often observed during daytime, suggesting influence from local vegetation.

Figure 6 shows the time series of CO₂ mixing ratio observed in four different altitude ranges in the FT and the surface pressure taken from the NCEP data for June 2007. In that year, the Pacific high was weak and the Baiu front was stalled between 20° and 30°N until mid-June. After the passage of a strong low-pressure system on 14–15 June, the Pacific high gained strength and the Baiu front often approached the Japanese main island, resulting in a number of cyclone passages over the region. During the latter half of the month, the fluctuation of CO₂ was much larger and the vertical distribution was much less uniform (Fig. 6). As can be seen in Fig. 6, the increase in CO₂ variability is more noticeable in the bottom half of the FT. High CO₂ concentrations were often observed in the warm

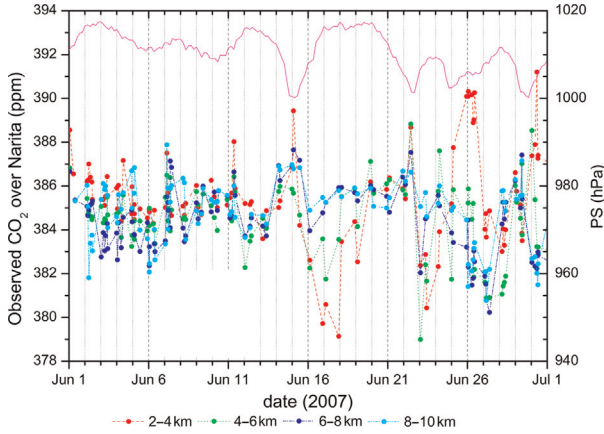


Fig. 6. Observed atmospheric CO₂ concentration at different altitudes (scale on the left: 2–4 km, 4–6 km, 6–8 km, 8–10 km) and surface pressure (scale on the right) over Narita in June 2007.

sector of a low-pressure system, while low CO₂ concentrations were observed in the cold sector after the passage of a cold front. This can be explained by the mechanism of mid-latitude cyclone airstreams reviewed by Stohl (2004). The surface emission from Asia is lifted into the atmosphere mainly by the warm conveyor belt (WCB), located on the eastern side of the cyclone. Meanwhile, in the north-easterly sector behind the cold front, the airmass transport bears the signature of an enhanced CO₂ uptake by boreal vegetation in summer. The above invocation of the transport mechanism by which pollution is vented from the emission sources in continental Asia to the North Pacific, called the ‘flushing effect’ by Sawa et al. (2007), provides a plausible explanation for the observation shown in Fig. 6, which is clearly supported by the simulated results shown in Fig. 7. The temporal evolution of the CO₂ concentration field associated with a strong cyclone passage from west to east is shown by the simulated changes in CO₂ at 700 hPa over East Asia/Japan (Fig. 7a); the corresponding movement of the synoptic systems from 13–16 June, 2007 is shown in Fig. 7b as daily weather charts obtained from the Japanese Meteorological Agency (JMA, 2007).

High and low events in the FT observed over Narita were caused mainly by the transport of the anthropogenic flux from East Asia (mainly China) and the TB flux from East Russia, respectively. The observed high SD of CO₂ in the summertime was caused by the synoptic fluctuation in the airmass, carrying high concentrations of anthropogenic CO₂ mainly from China and low concentration of CO₂ from the boreal forests in East Russia. As we can see in Fig. 2, the model often underestimated the low CO₂ deviation observed in the summer. This could be due to the underestimation of the CO₂ uptake by the boreal forests in East Russia.

3.3. Sensitivity to the regional fluxes and transport

To evaluate the relative influence of transport and flux variations on the CO₂ SD over Narita, a model sensitivity analysis was conducted. In this analysis, we essentially multiplied the emission values (fossil and vegetation) by two in all the tagged regions and investigated the response of the atmospheric CO₂ SD over Narita. The response of the SD is shown as % change from the reference CO₂ SD field over Narita (Fig. 8f). Any change (response) in the simulated CO₂ SD caused by perturbed forcing is presented as deviation from the reference CO₂ SD field. Thus, a small change in mixing ratio can produce a large percentage change in SD at a height with small SD, and vice versa. Multiplying the % change by the reference SD value (ppm) shown in Fig. 8f will give the ppm change caused by the doubling of the flux.

When the FF fluxes from East Asia and those from Japan were doubled, changes in the annual mean SD over Narita were found to be: 41 and 3% at 9 km, 61 and 4% at 5 km and 19 and 83% at 0.5 km, respectively. This result indicates that the SD over Narita is sensitive to transport (synoptic-scale meteorological variability) from upwind in the FT, but depends largely on the magnitude of local fluxes in the PBL.

Among 12 tracers, those that made more than 20% difference in monthly CO₂ SD were the FF fluxes from Japan and East Asia, and the TB fluxes from East Asia and East Russia. The impact on the CO₂ SD over Narita from changes in these sources is shown in the cross-sectional plots of altitude vs. time in Fig. 8. Fig. 8a and 8b indicate that the CO₂ SD in the PBL and in the FT are very sensitive to the anthropogenic flux from Japan and East Asia, respectively. Since the FF fluxes are constant throughout the year, any change in the SD produced by the model is mainly attributable to the variability in the transport and amplified by the increased CO₂ concentration in the atmosphere. The increase in the FF flux from Japan (Fig. 8a) results in an overall increase in the CO₂ SD in the PBL, with relatively little effect in the FT. The increase in the SD during July and August is caused by increased turbulent mixing in the PBL. In contrast, the doubling of the FF emission over the continental East Asia produces relatively little change in the CO₂ variability inside the PBL over Narita, but does show a significant increase of the CO₂ SD in the atmosphere above the PBL (Fig. 8b). It is interesting to note, however, a clear indication of a decrease in the mid-troposphere (from about 3 to 7 km) during the month of August; this is likely caused by the balance between the FF flux and the TB flux in the summer season. Increasing the atmospheric CO₂ concentration by doubling the FF emission over East Asia has a masking effect on the propagation of the negative deviation caused by the TB flux in the region in the summer.

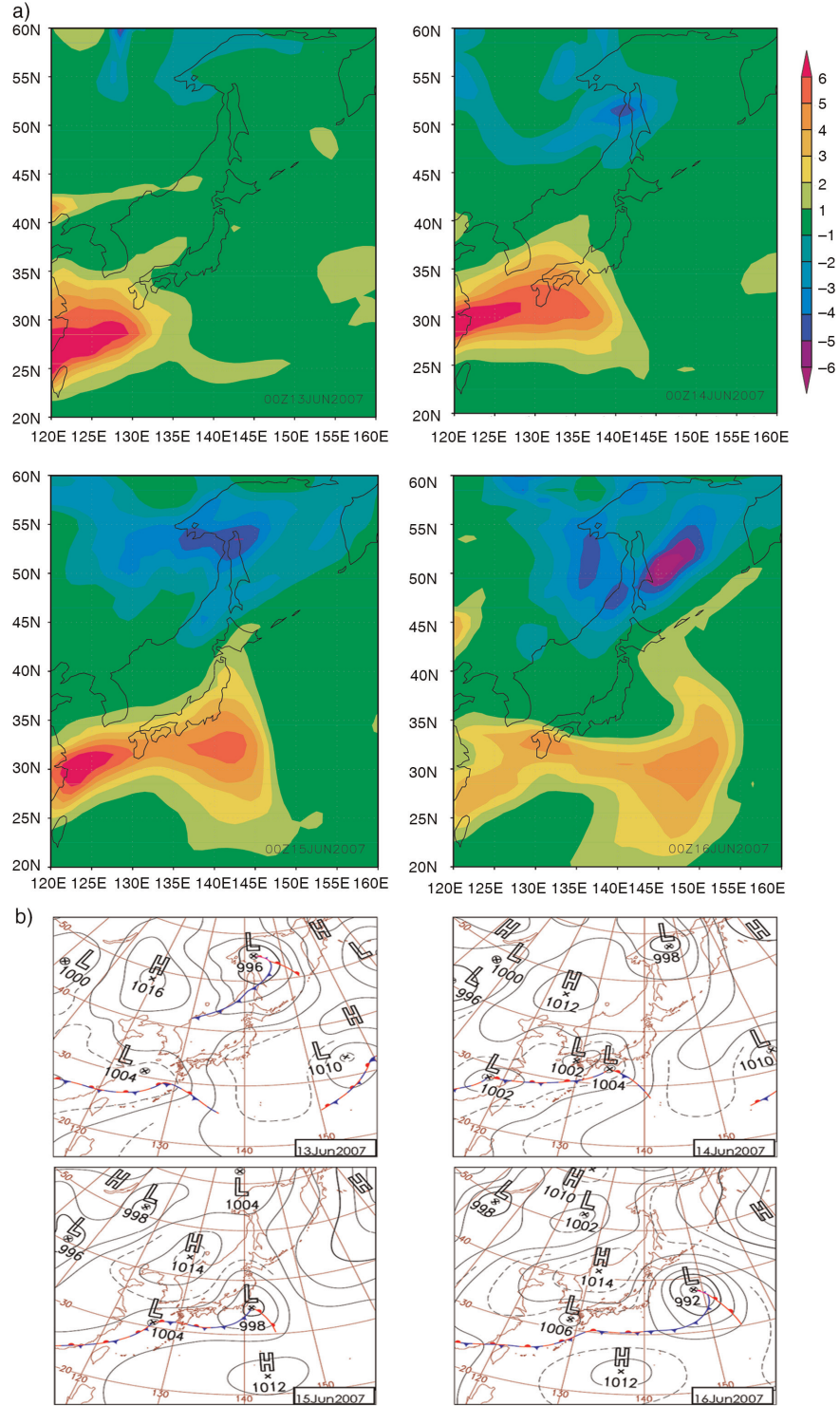


Fig. 7. Time series of a) CO₂ distributions in ppm (colour shaded contours) simulated by the model at the 700 hPa (≈ 3 km) altitude level and b) surface weather charts of East Asia and Japan at 00h UTC, from 13 to 16 June 2007. CO₂ values are standardised.

The TB flux from East Asia and East Russia affect the CO₂ SD in the FT in the summer, when strong CO₂ uptake makes large negative flux. Unlike the case involving changes

in the FF emission, the effect on the Narita SD of increasing the TB flux is more complicated to evaluate, since doubling the TB flux means doubling the emission/absorption

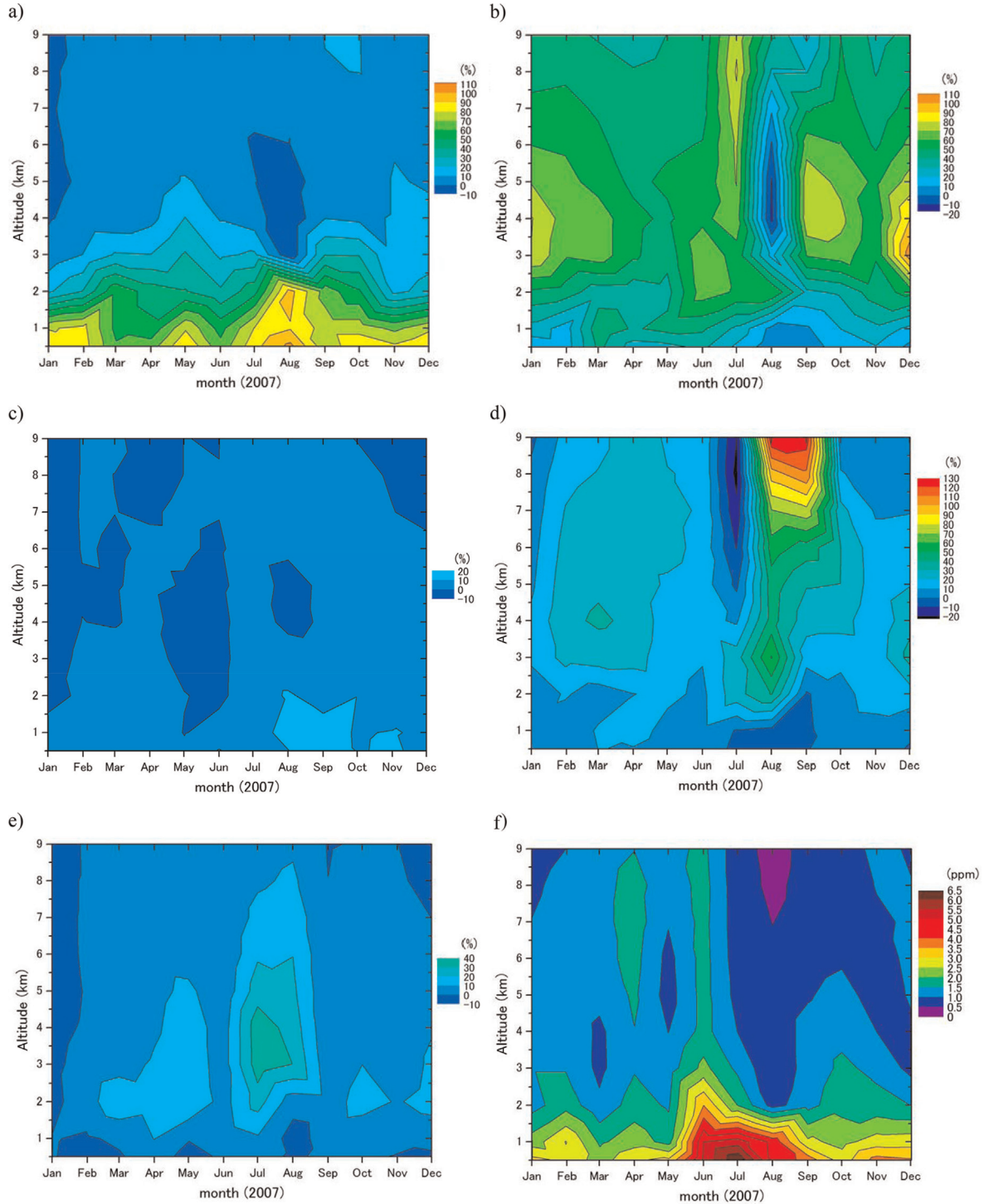


Fig. 8. Contour plots of changes (%) in monthly CO₂ standard deviation (SD) over Narita in response to a 100% increase of the anthropogenic flux from a) Japan and b) East Asia, and of the terrestrial biosphere (TB) flux from c) Japan, d) East Asia and e) East Russia. The contour plot of monthly CO₂ SDs (ppm) simulated with the standard flux set is shown in Fig. 8f. Altitudinal resolution is 1 km.

seasonal cycle. Doubling the amplitude of the seasonal TB flux from East Asia resulted in a decrease in the CO₂ SD over Narita above 4 km in July (with a decrease of 20% or

more in the upper troposphere). The variability, however, increased dramatically in the upper troposphere thereafter, reaching maximum values greater than 100% in August and

September (Fig. 8d). This is a result of the interplay between the magnified rapid downward shift from emission to absorption in the TB flux from late spring to early summer, the enhanced vertical transport of the surface flux signal to the upper troposphere [with a time delay of about 1–2 months (Nakazawa et al., 1993)] over East Asia, and the subsequent horizontal advection of this signal to Narita. The large percent values are also due to the very small net CO₂ SD in the upper troposphere in August and September (Fig. 8f), that makes it extremely sensitive to changes in the surface flux, which in this case is the TB flux from East Asia. The asymmetry in the magnitude of the change from relative decrease to increase in the CO₂ variability, compared to the standard model run, is likely due to the asymmetry of the TB seasonal flux. The impact of the TB flux from East Russia is shown in Fig. 8e. The increase in the Narita SD from this source shows up mainly in the 2–4 km layer in July and August. Compared to the influence of East Asia, the impact is relatively small, reaching a maximum increase of about 40%.

The result of the sensitivity study indicates that the synoptic-scale variability in the FT and in the PBL over Narita is most sensitive to changes in the anthropogenic and the TB fluxes in East Asia and to the anthropogenic flux in Japan, respectively. However, in order to interpret the result of this sensitivity study properly, the details of atmospheric transport modelling have to be investigated further. In the FT, model errors in diagnostic vertical wind are known to enhance vertical mixing and to cause a reduction in gradients and variability. In the PBL, numerical diffusion depends on the model resolution and tends to lower the variability of tracer mixing ratios (Book et al., 1991). Together with the uncertainty in advection due to wind errors, the uncertainties in mixing heights can be considerable (Lin and Gerbig, 2005; Gerbig et al., 2008). In addition, for Narita, which is located close to Tokyo and influenced by land-sea breeze circulations (Wakamatsu et al., 1983; Uno et al., 1984), representation errors are potentially significant. The latter issues should be improved by using a mesoscale Lagrangian model with finer flux distribution in the vicinity of the observation site.

Aircraft measurements can be more representative of a large-scale integrated regional surface flux than surface measurements that are often significantly influenced by local processes. A recent result of the CO₂ source/sink inversion using the CONTRAIL data indicated that fluxes for regions with strong vertical transport can be effectively constrained by the aircraft measurements (Niwa et al., 2012). This study demonstrated the sensitivity of the SD to the surface flux signals of upwind source/sink regions. These SD responses could potentially be used as additional constraints for regional inversions in the future.

4. Conclusions

The seasonal/vertical variation of SD of CO₂ is controlled by the combination of the regional flux distribution and of the atmospheric transport. It is noted that surface fluxes in upwind regions can impact the CO₂ distribution in the FT very quickly and directly around frontal zones via sporadic intrusion of PBL air. Over Narita, the SD was mostly affected by the CO₂ fluxes from East Asia in the FT and by fluxes in Japan in the PBL, respectively. Tagged simulation clearly showed enhanced transport of the CO₂ flux from East Asia to the upper troposphere over Narita in the spring, suggesting the passage of an active Asian outflow. This study is the first to clearly demonstrate the frequency of the Asian outflow in the spring based on the observed high CO₂ fluctuations detected in a multi-year record. A sensitivity analysis was performed to evaluate the relative influence of transport and flux variations on the CO₂ SD over Narita for 2007. When the regional fluxes that contributed the most, the FF fluxes from East Asia and those from Japan, were doubled, changes in the annual mean SD over Narita with height were 41 and 3% at 9 km, 61 and 4% at 5 km and 19 and 83% at 0.5 km, respectively. This result indicates that the SD over Narita is sensitive to transport (synoptic-scale meteorological variability) from upwind in the FT, but depends largely on the magnitude of local fluxes in the PBL. The seasonally-varying vertical profiles of atmospheric CO₂ SD over Narita showed height sensitivity to the CO₂ fluxes from different regions. Thus, inclusion of vertical profile information of atmospheric CO₂ variability could potentially provide a useful additional constraint to the inversion calculation of regional CO₂ fluxes. Since the SD is a non-linear function of concentration and flux, separating out the respective contribution of different sources is difficult, but the above results show at least qualitatively the usefulness of the information contained in the atmospheric CO₂ variability for estimating the CO₂ budget.

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