Seasonal and Long-Term Variations in Atmospheric CO₂ and ⁸⁵Kr in Tsukuba, Central Japan

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Abstract

Since February 1992, measurements of atmospheric CO₂ at 200 m above the ground have been taken from a meteorological tower (lat 36.1°N, long 140.1°E, 25 m a.s.l.) in Tsukuba, central Japan. We have examined seasonal/long-term variations in atmospheric CO₂, based on atmospheric ⁸⁵Kr, a noble gas with well-known source and sink terms, monitored continuously at weekly intervals in Tsukuba since May 1995. During the annual cycle, the daily mean values of atmospheric CO₂ in the afternoons (1300–1600 JST) showed maximum in late April, except in 2002, and minimum in late August or early September. The seasonal variations in atmospheric ⁸⁵Kr, which is low in summer and high in winter, were caused by the seasonal variations in the large-scale atmospheric circulation, and latitudinal distribution of atmospheric ⁸⁵Kr. Compared with seasonal variations in atmospheric ⁸⁵Kr, the draw-down (DD) and buildup (BU) for atmospheric CO₂ occurred about 1 week and 3 weeks later, respectively. The peak-to-trough amplitude of de-trended seasonal variations in atmospheric CO₂ concentrations ranged from 11 to 14 ppm. A decrease in the seasonal CO₂ minimum occurred with a decrease in the ⁸⁵Kr concentration, showing the effect of air mass transport from different origins. Unlike the minimum, the maximum CO₂ did not vary as expected from the air mass transport. The daily mean values of atmospheric CO₂ increased at an average rate of 2.0 ppm yr⁻¹, with a range from -0.5 to 4 ppm yr⁻¹. While both CO₂ and

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^{85}Kr are emitted into the atmosphere, due to energy consumption, the growth rate of atmospheric CO_2 showed a pattern different from that of atmospheric ^{85}Kr ($0.03 \text{ Bq m}^{-3} \text{ yr}^{-1}$).

1. Introduction

The CO_2 exchange between the atmosphere and the terrestrial biosphere is one of the important processes that determine the atmospheric CO_2 level currently increasing due to human activities. Although a number of studies of CO_2 uptake by the terrestrial biosphere have been conducted, estimates remain very uncertain (see for example, IPCC 2001). One way to decrease the uncertainty is to examine the atmosphere-biosphere CO_2 exchange, based on atmospheric measurements over land (Tans et al. 1996; Rayner et al. 1996). In order to estimate CO_2 exchange over such wide areas, it is necessary to determine the atmospheric CO_2 concentration on a regional scale, broadly defined as 10^4 to 10^6 km^2 (Helliker et al. 2004). To meet this requirement, tall towers have been used as monitoring sites since the 1990s (Bakwin et al. 1995, 1998, 2004; Haszpra 1999).

In February 1992, we started measuring atmospheric CO_2 at 200 m above the ground on a meteorological tower that stands in a field at the Meteorological Research Institute (MRI) (Fig. 1; MRI, lat 36.1°N , long 140.1°E , 25 m a.s.l.) in Tsukuba, Japan. Inoue and Matsueda (2001) reported that the daily mean values of atmospheric CO_2 measured from 1300 to 1600 Japan standard time (JST) showed maximum values in late April, and minimum values in late August to early September. The peak-to-trough amplitude of the de-trended seasonal variations in CO_2 concentrations ranged from 11 to 14 ppm. Year-to-year changes in the seasonal variations in atmospheric CO_2 could be caused by variations in photosynthesis/respiration of vegetation. In addition to variation in CO_2 flux, Dargaville et al. (2000) reported the effect of the interannual variability of atmospheric transport on atmospheric CO_2 observation. Higuchi et al. (2002) also demonstrated the importance of the need to use observed wind when carrying out interpretations of observed interannual atmospheric CO_2 variability, in terms of surface flux changes. It is important for a better understanding of the regional carbon cycle, to evaluate the contribu-

tion of each process to the year-to-year changes.

At the MRI, atmospheric ^{85}Kr activity concentration in ground level air has been monitored on a near weekly basis since May 1995 (Igarashi et al. 2000, 2001; Hirota et al. 2004). Levels of ^{85}Kr , a radioactive noble gas with a half-life of 10.8 years, have been increasing, mainly due to releases from nuclear fuel reprocessing plants. The only relevant sink for atmospheric ^{85}Kr is radioactive decay. After removing ^{85}Kr data collected during the operation of a nuclear reprocessing plant in Tokai, 60 km northeast of Tsukuba, the ^{85}Kr activity concentration showed a long-term increase and a clear seasonal variation, low in summer and high in

Fig. 1. The location of MRI in Tsukuba and PNC in Tokai. The upper panel shows the location of the monitoring site for atmospheric CO_2 in eastern Asia and the North Pacific. The lower panel shows the location of the area surrounded by the dashed line in the upper panel.

fall and winter (Igarashi et al. 2001). The well-known source/sink terms, and the chemically inert property of ^{85}Kr , allowed us to evaluate regional/long-range air mass transport and their relation to seasonal variations in atmospheric CO_2 .

In this paper, we report temporal variations in atmospheric CO_2 in Tsukuba from February 1992 through December 2003, along with that of atmospheric ^{85}Kr from May 1995 to December 2003, and discuss year-to-year changes in the seasonal and long-term variations of atmospheric CO_2 , based on ^{85}Kr concentrations.

2. Experiment

The site and measurements of atmospheric CO_2 , and ^{85}Kr were described in detail previously (Inoue and Matsueda 1996, 2001; Igarashi et al. 2000, 2001; Hirota et al. 2004). Therefore, only a brief description is given here.

2.1 Measurements of atmospheric CO_2

The MRI is located in Tsukuba, a city on the eastern part of the Kanto Plain, 50 km northeast of Tokyo, Japan (Fig. 1). There are vegetation, forests, and agricultural land in and around the urban area.

Air samples at 1.5, 25, and 200 m above the ground are continuously pumped at a rate of 5 to 10 L min^{-1} through copper tubes coated with polyvinyl chloride on the outside. There is a total of 500 m of copper tubing between the air intake at 200 m above the ground, and the system for measuring atmospheric CO_2 , which is located in the field experiment building. After removal of water vapor, the air sample is introduced at a rate of 0.3, or 0.4 L min^{-1} into a non-dispersive infrared gas analyzer (NDIR analyzer, Beckman model 865 or 880) placed in a plastic box to maintain a constant temperature in the analyzer cell. The NDIR analyzer is calibrated every 3 hours by successively introducing four calibrated working gases (330, 360, 390, and 420 ppm CO_2 in dry air) into the NDIR analyzer cell for 6 minutes each. The CO_2 concentration of working gas is traceable to the WMO mole fraction scale (Inoue and Matsueda 2001). Measurements of atmospheric CO_2 are taken every 6 minutes successively from the intakes at 25, 200, and 1.5 m, and continued in this order until the next calibration. Based on the replicate measurements of

a sample gas in the cylinder, the precision of analysis ($\pm 1\sigma$) was estimated to be better than ~ 0.1 ppm (Inoue and Matsueda 1996).

2.2 Measurement of atmospheric ^{85}Kr

The MRI has developed an atmospheric ^{85}Kr monitoring system through an international cooperation program with the Bundesamt für Strahlenschutz (BfS) Germany. Details of the BfS system are given in Stockburger et al. (1977). Samples of about 10 m^3 of air 15 m above the ground have been collected at the MRI on a near weekly basis since May 1, 1995. A specially designed metal adsorption vessel, filled with activated charcoal is immersed in liquid N_2 . The filtered air passes through the vessel at a flow rate of 1 L min^{-1} , and a low pressure of 0.5 atm, in order to avoid condensation of N_2 and O_2 . Water, and a large amount of CO_2 , are adsorbed on the metal fins at the upper part of the vessel, and krypton is quantitatively trapped on the activated charcoal. After passing through the sample air, the vessel is warmed to the ambient temperature until most of the remaining gases (N_2 , O_2 , CO_2 , etc.) and water vapor are removed from of the vessel. By heating the adsorption vessel to 350°C for 1 hour, and pressuring the vessel with He gas, krypton is transferred to an aluminum vessel (1 L) for sample transport to the BfS. The krypton is further purified at the BfS by gas chromatographic separation, using CH_4 as a carrier gas. The purified krypton fraction is introduced into a gas proportional counter with CH_4 , and its radioactivity is measured. After the end of the activity measurement the amount of stable Kr in the counter is determined by gas chromatography. The activity concentration of atmospheric ^{85}Kr is calculated, using the specific radioactivity ($^{85}\text{Kr}/\text{stable Kr}$) measured and the world wide constant abundance of stable krypton in the atmosphere (1.14 ppm, CRC 2000). The precision of the ^{85}Kr measurement was reported to be 1% (Weiss et al. 1992 and WMO 1996).

A nuclear fuel reprocessing plant is operated by the Power Reactor and Fuel Development Corporation (PNC, renamed the Japan Nuclear Cycle Development Institute in October 1998) at Tokai, about 60 km northeast of the MRI. The amount of ^{85}Kr emission at the Tokai plant is estimated to be a few percentage of the global

emission (United Nations 2000; Winger et al. 2005). Igarashi et al. (2000) reported that releases from this plant raised the ^{85}Kr level up to 15 Bq m^{-3} in weekly samples collected at the MRI in Tsukuba. In the current work, ^{85}Kr data observed during the operation of the Tokai plant were removed from the time series analysis. Therefore, ^{85}Kr data at the MRI can be used as a tracer of air mass with different origins, from which we can discuss the contribution of air mass to the variations in daily means of atmospheric CO_2 concentration.

2.3 Atmospheric CO_2 data at monitoring stations

Atmospheric CO_2 data from other monitoring sites, Ryori (RYO, lat 39.0°N , long 141.8°E , 230 m a.s.l.), Minamitorishima (MNM, lat 24.3°N , long 154.0°E , 8 m a.s.l.), Yonagunijima (YON, lat 24.5°N , long 123.0°E , 30 m a.s.l.), Shemya Island (SHM, lat 52.7°N , long 174.1°E , 40 m a.s.l.), Ulaan Uul (UUM, lat 44.5°N , 111.1°E , 914 m a.s.l.), Tae-ahn Peninsula (TAP, lat 36.7°N , long 126.1°E , 20 m a.s.l.), and Midway (MID, lat 28.2°N , long 177.2°W , 4 m a.s.l.) were used for comparison purposes. The first three sites are operated by the Japan Meteorological Agency (JMA), and the remaining four sites are part of the National Oceanic and Atmospheric Administration/Earth System Research Laboratory (former Climate Monitoring and Diagnostics Laboratory, NOAA/CMDL) flask-sampling network (Conway et al. 1994). The CO_2 data measured at the MRI are available from the World Meteorological Organization/World Data Center for Greenhouse Gases (<http://gaw.kishou.go.jp/wdcdgg.html>, WMO WDCGG) at the JMA, Tokyo, and those from other monitoring sites are available from the WMO WDCGG and Carbon Dioxide Information Analysis Center (<http://cdiac.esd.ornl.gov/index.html>) at Oak Ridge National Laboratory, Oak Ridge, TN, USA.

2.4 Time series analysis

The data of atmospheric CO_2 and ^{85}Kr were smoothed in time, using the methods described by Thoning et al. (1989):

$$f(t) = \sum_{i=0}^4 A_i t^i + \sum_{j=1}^h [B_j \cos(2\pi j t) + C_j \sin(2\pi j t)] \quad (1)$$

where t denotes the time in years since January 1, 1992, for atmospheric CO_2 , and since January 1, 1995 for atmospheric ^{85}Kr . We made the equally spaced data, with 1-week interval beginning on January 1, 1995, by taking the midday of sampling period for ^{85}Kr measurement into account. For this work, we used $h = 3$ for atmospheric CO_2 and $h = 2$ for atmospheric ^{85}Kr . The coefficients A_i , B_j , and C_j are constants determined by the least squares method. We removed CO_2 concentration data located from the curve by more than two times the residual standard deviation ($\text{RSD} = 7.0 \text{ ppm}$ for atmospheric CO_2 , $\text{RSD} = 0.04 \text{ Bq m}^{-3}$ for atmospheric ^{85}Kr). The remaining data were again fitted to Eq. (1) by the least squares method. We repeated the data selection procedure twice for atmospheric CO_2 . We then calculated the residuals of the retained data from values obtained by Eq. (1), and smoothed them by a low-pass filter with a full width at a half maximum of about 100 days. Before the application of the low-pass filter, the continuous residual data were extended by 365 days at each end of the series by repeating the first and last years of data (Thoning et al. 1989). The residuals smoothed with the low-pass filter were added to $f(t)$ to show variations on a time scale longer than a few months ($\text{RSD} = 3.9 \text{ ppm}$ for atmospheric CO_2 , $\text{RSD} = 0.03 \text{ Bq m}^{-3}$ for atmospheric ^{85}Kr). In order to analyze the long-term trend, a low-pass filter with a full width at a half maximum of about 667 days was applied to the data. The statistical uncertainty associated with each value in the filtered data was calculated to be 0.4 ppm for CO_2 , and 0.01 Bq m^{-3} for ^{85}Kr (Thoning et al. 1989).

3. Results and discussion

3.1 Long-term variations in atmospheric CO_2 and ^{85}Kr

Atmospheric CO_2 at 200 m above the ground in Tsukuba clearly shows that a large diurnal variation occurs in summer, and a smaller one occurs in winter (Inoue and Matsueda 2001). In summer, the maximum occurs a few hours after dawn and the minimum in the afternoon. The maximum is caused by the convection which transports upward air with high CO_2 concentration from the nocturnal stable layer. The minimum is caused by the photosynthesis

Fig. 2. Temporal variations in daily means of selected atmospheric CO_2 data at 200 m above the ground from February 1992 through December 2003. The top panel (A) shows the selected data used for curve fitting. The middle panel (B) shows the smooth curve fitting to daily mean CO_2 concentrations (solid line) and the long-term trend (thin line). The bottom panel (C) shows the growth rate of atmospheric CO_2 .

of land plants and the active vertical mixing of air. In winter, the CO_2 concentration is fairly constant because of temperature inversion below 200 m. In this study, we used daily mean values of atmospheric CO_2 in the afternoon (1300–1600 JST), which were expected to be close to representative values in the planetary boundary layer. Figure 2 illustrates temporal variations in the daily means of selected atmo-

spheric CO_2 data from February 1992 to December 2003.

The atmospheric CO_2 concentration increased at a mean rate of 2.0 ppm yr^{-1} , varying from -0.5 to 4 ppm yr^{-1} (Fig. 2). In Tsukuba, a relatively high growth rate of atmospheric CO_2 occurred in late-1997/early-1998, that coincided with a large global average growth rate for CO_2 (Conway et al. 1994; <http://www.cmdl.noaa.gov/ccg/index.html>). In Tsukuba a low growth rate occurred in the middle of 1994, but global average growth rate did not decrease at that time. Superimposed on the variations in the global average growth rate for CO_2 , are variations on relatively small spatial scales.

The ^{85}Kr activity concentration increased at an average growth rate of $0.03 \text{ Bq m}^{-3} \text{ yr}^{-1}$ from 1995 to 2003 (Fig. 3), where the relative increase rate ($2\% \text{ yr}^{-1}$) was much faster than that of atmospheric CO_2 ($0.4\% \text{ yr}^{-1}$). Both CO_2 and ^{85}Kr are emitted into the atmosphere mainly because of energy consumption. However, the growth rate of atmospheric ^{85}Kr shows a pattern different from that of atmospheric CO_2 . ^{85}Kr is released into the atmosphere during the reprocessing of used nuclear fission fuels, and no relevant sinks exist other than radioactive decay. Hirota et al. (2004) reported that annual averages of ^{85}Kr concentration in Tsukuba were in good agreement with those calculated based on the release rate of ^{85}Kr from the nuclear fuel reprocessing plants in Europe. Large interannual variations in ^{85}Kr emission (Winger et al. 2005) after the use of energy, and variations in source/sink strengths for CO_2 , probably lead to the different pattern of growth rate for the two gases.

3.2 Seasonal variations in atmospheric CO_2 and ^{85}Kr

Except in 2002, atmospheric CO_2 had seasonal variations similar to those observed at baseline stations in northern mid-latitudes (Conway et al. 1988). The CO_2 concentration reached a maximum in April and a minimum in late August or early September (Fig. 2). In 2002, the atmospheric CO_2 concentration reached maximum in February. The difference in CO_2 concentration between 1.5 and 200 m above the surface was small (0.1 ppm), as compared with those of the other years (0.9 – 2 ppm), due to a larger increase in CO_2 concen-

Fig. 3. Temporal variations of selected atmospheric ^{85}Kr data in Tsukuba from May 1995 through December 2003. The top panel (A) shows the selected data used for curve fitting. The middle panel (B) shows the smooth curve fitting to selected ^{85}Kr concentrations (solid line) and the long-term trend (thin line). The bottom panel (C) shows the growth rate of atmospheric ^{85}Kr .

tration at 200 m. The enhanced vertical mixing led to the maximum CO_2 concentration in February 2002. The ^{85}Kr activity concentrations observed in Tsukuba clearly showed seasonal variations (Fig. 3). The seasonal minimum occurred in early August, and the maximum occurred in late November or December. Seasonal variations in atmospheric ^{85}Kr are caused by seasonal variations in the large-scale atmospheric circulation, and the latitudinal distri-

bution of atmospheric ^{85}Kr (Igarashi et al. 2000). Levin and Hesshaimer (1996) reported that the maximum ^{85}Kr occurred slightly south of 60°N ; furthermore, they showed that the ^{85}Kr concentration decreased rather steeply south of the maximum, while decreasing gradually toward the North Pole (Weiss et al. 1992 and Winger et al. 2005). During summer, maritime air mass formed at lower latitudes hovers over Japan, while during winter, a continental air mass formed at higher latitudes dominates over Japan. The de-trended seasonal variations in ^{85}Kr concentrations show that an exchange of air masses, with different origins occurs fairly rapidly from May to July, and from September to October.

A possible cause of the maximum ^{85}Kr is the role of air mass transport at high latitudes where the ^{85}Kr concentration was high, or there was an enhanced release of ^{85}Kr from nuclear reprocessing plants. The monthly emission data of ^{85}Kr are available only from the nuclear reprocessing plant in La Hague, France (lat 49.0°N , long 0.9°W), the strongest source in the 1990s (Winger et al. 2005). According to the data, monthly emissions of ^{85}Kr in November and December were fairly equal to those in October and January (data not shown). If the air masses originating at latitudes higher than 60°N were advected to Japan during winter (January and February), ^{85}Kr concentrations that were relatively low as compared with that of 60°N , but higher than those at low latitudes could be expected. Hirota et al. (2004) reported a good correlation between ^{85}Kr concentration and the origin of an air mass estimated by backward trajectory analysis for 240 hours starting at an altitude of 1500 m. They reported that relatively low ^{85}Kr concentration could occur in January/February, due to air masses passing over the lower latitudes and the North Pacific (Fig. 6 in Hirota et al. 2004).

De-trended seasonal variations in atmospheric CO_2 and ^{85}Kr were calculated by subtracting values of the long-term trend from the seasonal values shown in Fig. 2. The CO_2 increased continuously during the winter season due to the accumulation of respired CO_2 from the land biosphere, and anthropogenic CO_2 , while atmospheric ^{85}Kr concentration maximum occurred in December. Table 1 contains a list of the maximum and minimum values of

Table 1. Seasonal variations in atmospheric ^{85}Kr in Tsukuba and CO_2 measured 200 m above the ground at the meteorological tower.

Year	Peak-to-trough amplitude		Maximum		Minimum		DD		BU	
	CO ₂ (ppm)	⁸⁵ Kr (Bq m ⁻³)	CO ₂ (ppm)	⁸⁵ Kr (Bq m ⁻³)	CO ₂ (ppm)	⁸⁵ Kr (Bq m ⁻³)	CO ₂	⁸⁵ Kr	CO ₂	⁸⁵ Kr
1995	11.2	0.17	4.7	0.06	-6.5	-0.11	3 June	21–27 May	26 October	1–7 October
1996	13.7	0.12	4.8	0.05	-8.9	-0.07	6 June	19–25 May	25 October	29 September–5 October
1997	11.9	0.15	4.8	0.07	-7.1	-0.08	29 May	15–21 June	1 November	12–18 October
1998	13.0	0.16	5.6	0.07	-7.4	-0.09	4 June	17–23 May	28 October	18–24 October
1999	11.4	0.17	5.0	0.06	-6.4	-0.11	3 June	30 May–5 June	8 November	3–9 October
2000	11.6	0.16	5.0	0.07	-6.6	-0.09	29 May	21–27 May	2 November	1–7 October
2001	13.2	0.16	4.8	0.07	-8.4	-0.09	9 June	13–19 May	31 October	7–13 October
2002	10.9	0.18	4.9	0.07	-6.0	-0.11	25 May	26 May–1 June	27 October	29 September–5 October
2003	14.0	0.13	6.2	0.06	-7.8	-0.07	4 June	19–25 May	27 October	18–24 October

the de-trended seasonal variations. To determine the phase of seasonal variation, Conway et al. (1988) examined the days when the value of the de-trended seasonal variations in atmospheric CO_2 concentrations became 0 ppm. These days were used as indicators of the phasing of CO_2 draw-down (DD) and CO_2 buildup (BU), respectively. Figure 4 shows the average feature of the de-trended seasonal variations in atmospheric CO_2 and ^{85}Kr from 1995 to 2003. The present records indicate that the average DD for atmospheric CO_2 occurred about 1 week later than that for atmospheric ^{85}Kr and the average BU for atmospheric CO_2 occurred about 3 weeks later. The relatively longer period between DD and BU for CO_2 could be caused by regional footprint (photosynthetic activity), or seasonally varying north-south gradient for CO_2 .

With respect to year-to-year changes in DD and BU, the DD for atmospheric CO_2 occurred from 29 May to 9 June, and the BU for atmospheric CO_2 occurred about 22 weeks later, from 25 October to 10 November (Table 1). The DD for atmospheric ^{85}Kr occurred from 13–19 May to 15–21 June (atmospheric ^{85}Kr was monitored on a near weekly basis), and the BU for atmospheric ^{85}Kr occurred about 20 weeks later, from 29 September–5 October to 18–24 October (Table 1). With respect to the phase of seasonal variations, DD (BU) for atmospheric CO_2 varied independently of DD (BU) for atmospheric ^{85}Kr . These results show that the phase of the seasonal variation in atmospheric CO_2 is not explicitly related to the air mass transport.

Peak-to-trough amplitudes of the seasonal variations ranged from 11 to 14 ppm for atmospheric CO_2 , and 0.12 to 0.18 Bq m^{-3} for ^{85}Kr (Table 1). The former was caused by the transport of air masses with different origins and/or CO_2 exchange between the air and the land biosphere in the footprint region. We examined year-to-year changes in the seasonal CO_2 minimum concentration against ^{85}Kr concentration at the time when the minimum CO_2 occurred (Fig. 5). The seasonal CO_2 minimum concentration decreased with a decrease in the ^{85}Kr concentration. Data collected at the sites shown in Fig. 1 indicated that atmospheric CO_2 concentrations at high latitudes were lower than those at low latitudes, due to the effect of photosynthetic activity on land from July to August.

Fig. 4. Averages of the de-trended seasonal variations in atmospheric CO_2 (solid line) and ^{85}Kr (dotted line) from 1995 through 2003.

Fig. 5. Relationship between the minimum value of the de-trended seasonal variations in atmospheric CO_2 ($[\text{CO}_2]_{\min}$) and ^{85}Kr concentrations ($[^{85}\text{Kr}]$) at the time when the minimum atmospheric CO_2 concentration occurred. The solid line shows the linear relationship ($[\text{CO}_2]_{\min} = -50.6(\pm 14.2) \times [^{85}\text{Kr}] - 10.8(\pm 1.0)$, $n = 9$, $p < 0.01$, $r = 0.803$, $1\sigma = 0.6$ ppm).

Therefore, the air masses from high latitudes had relatively low CO_2 concentrations and high ^{85}Kr concentrations.

Inoue and Matsueda (2001) examined year-to-year changes in CO_2 concentrations (ΔCO_2) over a fixed period when atmospheric CO_2 concentrations varied rapidly and monotonously. From 1 May to 31 July, the calculated ΔCO_2 increased (absolute value of ΔCO_2 decreased) with an increase in surface temperature in Tsu-

Fig. 6. Relationship between the average of monthly mean temperature (T_{av}) of 16 meteorological stations located within 160 km from Tsukuba in August (<http://www.data.kishou.go.jp>) and the minimum value of de-trended seasonal variations in atmospheric CO_2 ($[\text{CO}_2]_{\min}$). The solid line shows the linear relationship ($[\text{CO}_2]_{\min} = 0.66(\pm 0.11) \times T_{\text{av}} - 24.1(\pm 2.9)$, $n = 12$, $p < 0.001$, $r = 0.880$, $1\sigma = 0.5$ ppm).

kuba. From the viewpoint of CO_2 exchange between the atmosphere and land biosphere, the year-to-year changes could be explained by the regional footprint: either a decrease in photosynthesis or an increase in respiration. By taking into account meteorological parameters affecting biological activity, it can be inferred that the changes were caused by the latter because of a larger temperature dependence of respiration compared with that of photosynthesis (Inoue and Matsueda 2001). Figure 6 shows the relationship between the CO_2 minimum and the average of monthly mean temperature (T_{av}) of Kanto district in August. The CO_2 minimum increased with an increase in T_{av} in August. The higher T_{av} means the air mass originated from lower latitudes (usually maritime air) with higher CO_2 concentration. Therefore, both biological activity over Japan and air mass transport with different origins could lead to the observed relationship between the minimum CO_2 and ^{85}Kr concentration as long as the anthropogenic addition of CO_2 remained constant in central Japan from 1995 to 2003.

In April, the month when the atmospheric CO_2 maximum usually occurs in Tsukuba, the

Fig. 7. Relationship between the maximum value of the de-trended seasonal variations in atmospheric CO_2 and ^{85}Kr concentrations at the time when the atmospheric CO_2 maximum occurred.

CO_2 concentration increases from low to mid/high latitudes (GLOBALVIEW- CO_2 2004). Therefore, the maximum value of atmospheric CO_2 was expected to increase with the ^{85}Kr concentration if air mass transport played an important role in determining year-to-year changes in the CO_2 maximum in spring. However, the maximum value of atmospheric CO_2 did not increase with ^{85}Kr concentration (Fig. 7). The low correlation between the two may indicate a relatively minor role of air mass transport in determining the interannual variations in maximum CO_2 . Year-to-year variation in maximum CO_2 , ranging from 4.8 to 6.1 ppm, was small as compared with that of the minimum. We need detailed information on the spatial distributions of atmospheric CO_2 in the footprint region.

The atmospheric ^{85}Kr shows an air mass feature over central Japan, which can be used to discuss year-to-year changes in seasonal variations of atmospheric CO_2 . The data from these measurements are valuable for examining the effects of air mass transport from different origins, and CO_2 exchange between the atmosphere and land biosphere on the CO_2 concentration. However, in order to know from where and to what extent CO_2 exchange between the atmosphere and land biosphere affects CO_2 data, it is necessary to develop an integrated system of measurements, including chemical tracers which show sources/sinks (Bakwin et al. 1995; Gloor et al. 2001; Schmidt et al. 2003),

and atmospheric transport models (Dargaville et al. 2000; Higuchi et al. 2002; Taguchi et al. 2003; Murayama et al. 2004) for interpreting the temporal variations in atmospheric CO_2 in central Japan.

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