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Temporal and spatial variations of upper tropospheric and lower stratospheric carbon dioxide

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ABSTRACT

Systematic collection of air samples was made using commercial jet airliners between Tokyo, Japan and Anchorage, Alaska and between Tokyo and Sydney, Australia in 1984 and 1985. The amplitude of the seasonal CO_2 cycle in the upper troposphere was found to be largest in high latitudes of the northern hemisphere but decreased and lagged in phase as the equator was approached. The cycle was still clearly observable in the southern hemisphere, due to southward transport of the northern hemispheric air through the upper troposphere by a monsoon circulation. The yearly mean value of the upper tropospheric CO_2 was high in the equatorial region and decreased poleward. The seasonal cycle of the lower stratospheric CO_2 in northern high latitudes showed a minimum concentration early in May and a maximum concentration early in September, with a peak-to-peak amplitude of 2.2 ppmv. Yearly mean CO_2 concentrations were lower by 1.4–2.2 ppmv than those in the upper troposphere. The difference in yearly mean CO_2 concentrations between 1984 and 1985 was about 1.1 ppmv at all locations covered by this measurement.

1. Introduction

The concentration of atmospheric CO_2 has steadily increased over the last 200 years (Neftel et al., 1985) due to human activities such as fossil fuel combustion and deforestation. This increase will very likely enhance the greenhouse effect of the terrestrial atmosphere, leading to global climatic change.

One of the most important subjects for this problem is to elucidate the carbon cycle on the earth's surface. This is crucial for predicting future levels of the atmospheric CO_2 concentration. Substantial efforts have therefore been devoted to establishing a comprehensive network for background CO_2 monitoring. The present global network consists mainly of about 40 ground-fixed stations. The data from aircraft measurements are especially useful for developing and validating carbon cycle models (Pearman and Hyson, 1980, 1986; Fung et al., 1983; Gillette and Box, 1986; Heimann and Keeling, 1986; Keeling and Heimann, 1986; Tans et al., 1989), but they are very sparse. Systematic measurements with aircraft have been made over northern high latitudes, the Pacific Ocean, Japan and Australia (Keeling et al., 1968; Bolin and Bischof, 1970; Bischof, 1985; Pearman and Garratt, 1973; Pearman and Beardsmore, 1984; Tanaka et al., 1983a, 1987a, 1988). At present, only the programs over Japan and Australia are still operating. During the period from January 1984 to December 1985, we collected air samples on board commercial jet airliners between Tokyo, Japan and Anchorage, Alaska and between Tokyo and Sydney, Australia.

In this paper, we report temporal and spatial variations of the upper tropospheric and lower stratospheric CO_2 over a wide geographical area. The results are also compared with those of the lower tropospheric CO_2 which were obtained mainly from our other CO_2 programs.

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2. Experimental procedures

Details of our experimental procedures have been described elsewhere (Tanaka et al., 1983a, b, 1987a, 1988). Therefore, only brief description and supplemental explanations of the procedures are given here.

The air sampling was made once per month on a roundtrip flight between Tokyo and Anchorage and between Tokyo and Sydney. The aircraft used was a Boeing B747 and its flight routes are shown in Fig. 1. Air samples were collected from altitudes of approximately 10–12 km at intervals of 5° between 40°N and 60°N and at about 7.2 km height over Anchorage. On flights to Australia, samples were also collected from about 10–12 km at intervals of 5° between 30°N and 30°S and at 7.2 km over Sydney. However, as a total of only 20 sample flasks were available, the collection of samples on the return flights was limited to 5 positions at every 5° between 20°S and the equator from January 1984 to April 1985 and with every 10° between 25°S and 15°N for the remaining period.

Each air sample was taken from the so-called "fresh air" nozzles of the cockpit and pressurized into a 350 ml stainless-steel flask to an overpressure of about 3 atm with a manual stainless-steel-plunger pump. As mentioned by Pearman and Beardsmore (1984), the B747 aircraft are usually equipped with recycle fans between the engines and the nozzles to reduce fuel consumption by mixing the cabin air with the fresh air. However, in the aircraft used here, the fresh air compressed in the engines is brought to the nozzles of the cockpit without passing through the fans.

The sample flasks were cylindrical in shape with two O-ring seal stopcocks at the ends. After fabrication, they were washed sufficiently in a diluted acid solution and pure water activated by an ultra-sonic wave and then dried out by flowing

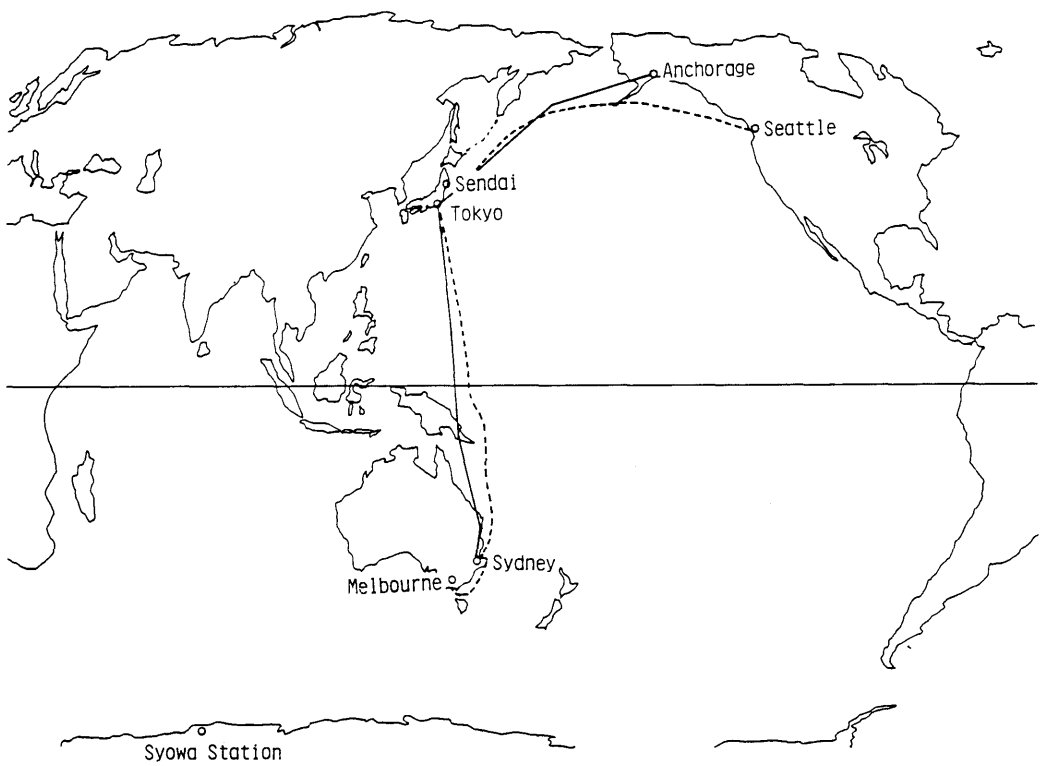

Fig. 1. Sampling routes for aircraft (solid lines) and ship-board (broken lines) measurements. Locations of Sendai and Syowa Station are also shown.

dry nitrogen gas through them. All flasks were heated at about 100°C and evacuated to lower than 1.3×10^{-3} Pa for 3 hours prior to use.

A possible deterioration of air samples during their storage in the flasks is a critical problem in determining precise values of the CO_2 concentration. We therefore examined the CO_2 concentrations of standard gases stored in the flasks. The results showed that the deterioration risk of air samples is negligible, concentration changes being within 0.1 ppmv for at least one week.

Air samples collected were returned to our laboratory and their CO_2 concentrations were determined using a non-dispersive infrared analyzer with a precision of 0.01 ppmv against our CO_2-in-air standard gases. This was done within 1 week after collection. Water vapor in air samples was removed by 2 glass traps cooled at −78°C. Therefore, the concentration values reported here are expressed in parts per million (ppm) by volume of dry air. Our standard gases were classified into 3 categories; primary, secondary and working. The concentrations of working standard gases were determined before and after their use with a precision of 0.01 ppmv by comparing them with our secondary standard gas system to confirm that any drifts in concentration were less than 0.1 ppmv. The secondary standard gases were compared at least twice each year with primary standards prepared gravimetrically with an uncertainty of 0.13 ppmv. The overall precision of our flask analysis was 0.1 ppmv.

3. Data analysis

About 640 air samples were collected and analyzed for 2 years. Visual inspection of the data showed that the CO_2 concentrations of air samples from 3 flights between Tokyo and Sydney are horizontally irregular and higher by about 5.0 ppmv than expected from the results of contiguous flights. Since the flow rate of the air from the nozzles was very low during these flights, it is thought that contaminated cabin air intruded into the sample flasks. Some other samples also showed high CO_2 concentrations due to a failure in sampling. These data were first excluded from the data set. The remaining data were grouped into the troposphere and the stratosphere, in the light of radiosonde observations. In this procedure, it was occasionally difficult to determine whether the sample was from the troposphere or not. It was also found that some of the upper tropospheric data taken between Tokyo and Anchorage indicated CO_2 concentrations lying between the tropospheric and stratospheric values, due presumably to the mixing of the two air masses by a tropopause folding (Tanaka et al., 1983a). Such data were not included in this data process. As a result, all of the data taken between Tokyo and Sydney and at 7.2 km height over Anchorage and Sydney were assigned to the troposphere, and about 70% of the data between Tokyo and Anchorage, to the stratosphere. A complete set of the upper tropospheric data was not obtained for every 5° between 40° and 60°N, because the tropopause height was lowered in high latitudes, especially from autumn to spring through winter.

The stratospheric data in the same month were simply averaged to calculate monthly mean values of the CO_2 concentration. The tropospheric data taken at the same location on each round-trip flight were also averaged. Values of the CO_2 concentration, thus obtained, showed clear evidence of the seasonal cycle and the trend. To separate respective contributions, a digital-filtering technique including Fourier harmonics, Reinsch-type cubic spline (Reinsch, 1967) and Butterworth filter (Hamming, 1977) was used in this study. Since details of this filtering process will be given elsewhere, its outline is described here.

We first fitted the observed data to the Fourier harmonics,

$$S(t) = \sum_{j=1}^{k} [A_j \sin(2\pi jt) + B_j \cos(2\pi jt)], \qquad (1)$$

and Reinsch-type spline with a cutoff frequency of 0.2 cycles/year at which a signal is 50% attenuated by the spline-fitting process (Cox, 1983; Enting, 1987). Here, t is the elapsed time (years) relative to 1 January 1984 and A_j's and B_j's are parameters determined by the fitting. The value of k was set to 2 and 3 to approximate the average seasonal cycles of the northern and southern hemispheric CO_2, respectively. As described below, the seasonal CO_2 cycle was more complicated in the southern hemisphere than in the northern hemisphere. Spectral analysis of the data by a maximum entropy method (Burg, 1967) also supported this finding, as shown in Fig. 2.

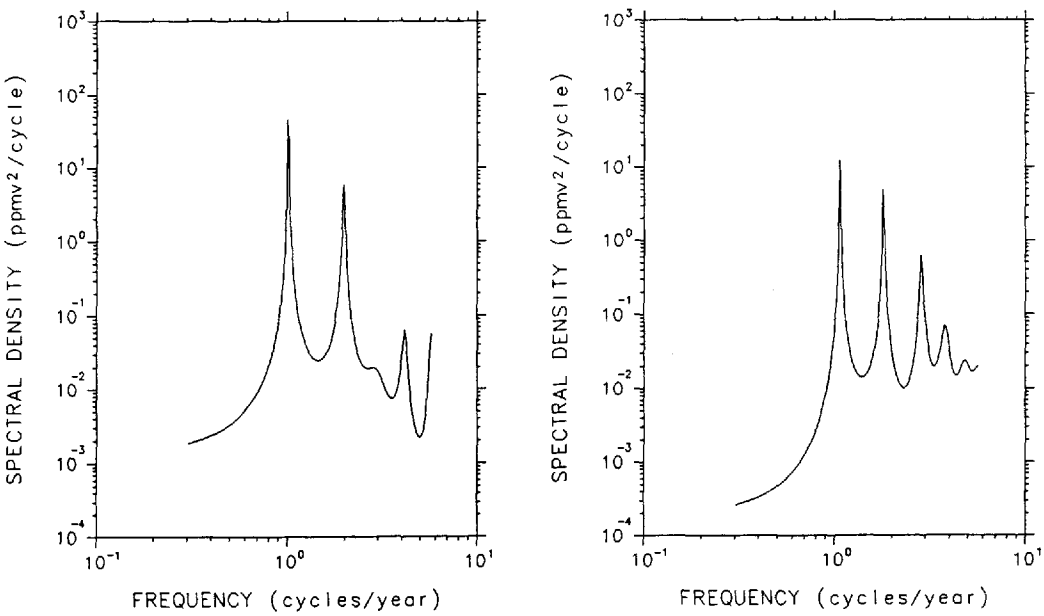

Fig. 2. Power spectra of the upper tropospheric CO_2 concentrations at 30°N (left) and 30°S (right).

The trend and the average seasonal cycle were determined by using an iterative procedure, after subtracting the average cycle approximated using eq. (1) from the data and adding the trend expressed by the above spline to the beginning and the end of the record by 3 and 8 years, respectively.

(1) The data record thus obtained was interpolated by the Reinsch-type spline to calculate daily values of the CO_2 concentration. Here, the cutoff frequency of the spline was chosen to be 7.3 and 9.1 cycles/year, for $k = 2$ and 3, respectively. These splines pass, with negligible attenuation, all signals with periods greater than the period at which the seasonal cycle, including irregular parts, is defined.

(2) Daily values of the CO_2 concentration were smoothed by the 26th-order Butterworth filter. To compensate for the phase shift occurred in the filtering process, its output was reversed and then passed through the filter again. The signal with a frequency of 0.5 cycles/year was attenuated to 50% by this procedure.

(3) The trend obtained in the above step was subtracted from the data and eq. (1) was fitted to residuals. The average seasonal cycle thus obtained was removed from the data, and the trend given initially or in previous iteration on both ends of the record was altered by the results of the above filtering process, so that actual data were followed as much as possible.

The steps (1)–(3) were repeated until the trend becomes unchangeable, which usually converged after a few iterations. Once the trend and the average seasonal cycle were determined, these components were removed from the data and zero values were added to the respective ends of the record by the same years as above. Then, the steps (1)–(3) were repeated again to derive irregular parts of the seasonal cycle. In this case, we adopted the 16th-order Butterworth filters to attenuate signals with periods of 4 and 3 months, to 50% for $k = 2$ and 3, respectively.

4. Results and discussion

The CO_2 concentrations observed in this measurement are plotted in Fig. 3, together with the results of our aircraft measurements made using a McDonell Douglas DC-9 between 8 km height and the tropopause over Japan for the same period (Tanaka et al., 1987a). Also shown are the

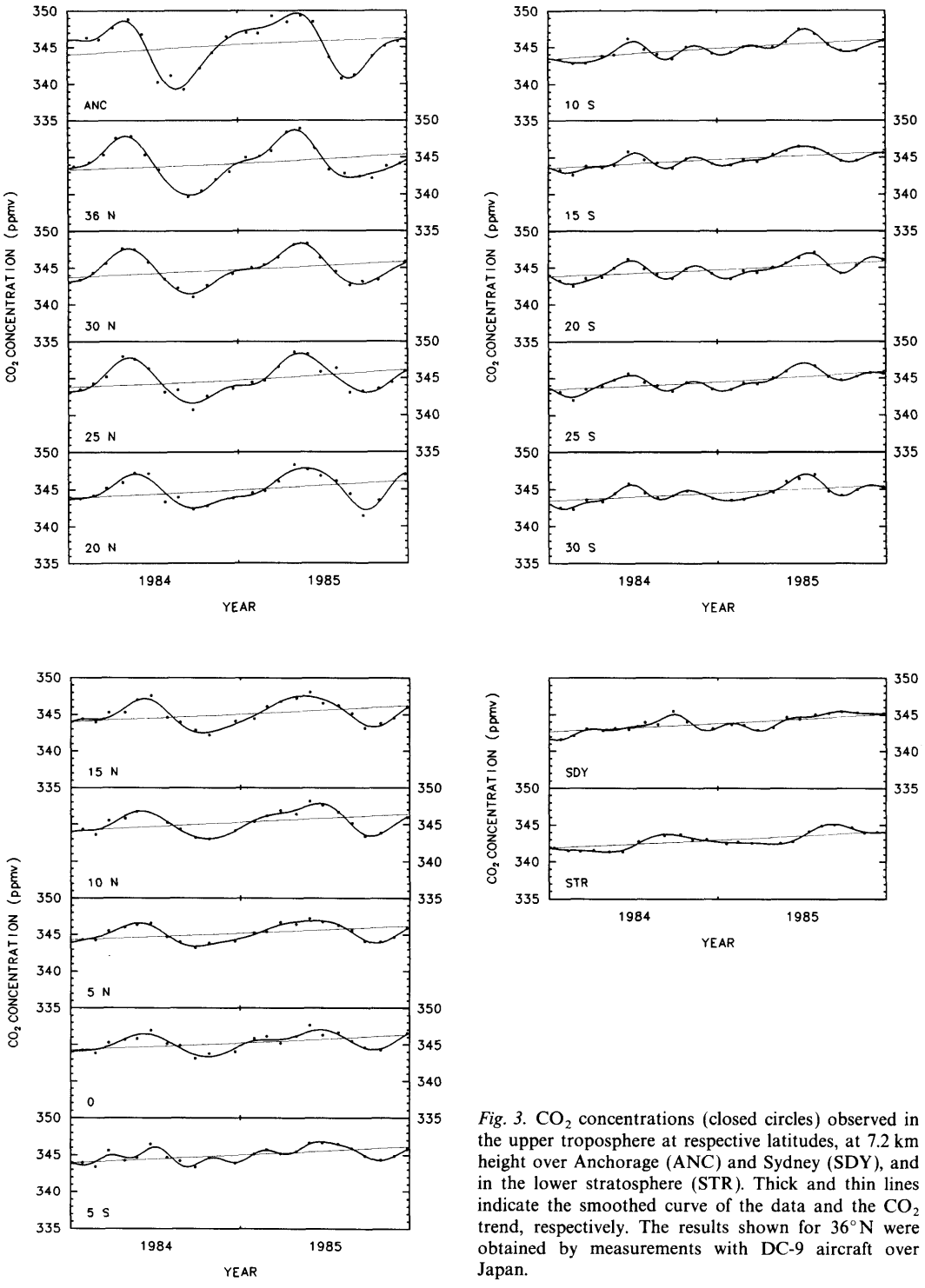

Fig. 3. CO_2 concentrations (closed circles) observed in the upper troposphere at respective latitudes, at 7.2 km height over Anchorage (ANC) and Sydney (SDY), and in the lower stratosphere (STR). Thick and thin lines indicate the smoothed curve of the data and the CO_2 trend, respectively. The results shown for 36°N were obtained by measurements with DC-9 aircraft over Japan.

CO_2 trends and smoothed curves of the data obtained by the above-mentioned digital-filtering technique. The standard deviations of observed values from the smoothed curves are listed in Table 1. As seen in Fig. 3, the concentration of atmospheric CO_2, with clear seasonal cycle, increased almost linearly at all locations.

Yearly mean values of the CO_2 concentration derived from the smoothed curves of the data are shown in Fig. 4 and Table 1. The upper tropospheric values indicate nearly the same latitudinal distribution in 1984 and 1985, showing an average concentration difference of about 1.1 ppmv between the two years; the value is high in the equatorial region centered at 5°N and decreases gradually poleward. Concentration differences between the equatorial region and middle latitudes of the northern and southern hemispheres are about 0.8 ppmv. Upward transport of the lower tropospheric air near the Intertropical Convergence Zone (ITCZ) may be responsible for this distribution. The ITCZ is usually located at 5°N on the Pacific Ocean to the windward of our flight routes. The upper tropospheric values at 36°, 30° and 25°N in 1984 are in good agreement with those of aircraft measurements with a Boeing B737 over the Japanese islands (Tanaka et al., 1988). Yearly mean CO_2 concentrations at 7.2 km height over Sydney are lower by about 0.7 ppmv than the upper tropospheric values at 30°S, and those over Anchorage are higher by about 0.7 ppmv than the upper tropospheric values over Japan. Our aircraft measurements over Japan in 1984 and 1985 showed that an average difference of yearly mean CO_2 concentrations between the uppermost part of the troposphere and 7 km height is about 0.3 ppmv. Therefore, the values over Anchorage are higher by 0.4 ppmv than those at the same altitude over Japan, due probably to upward transport of the lower tropospheric air in northern high latitudes.

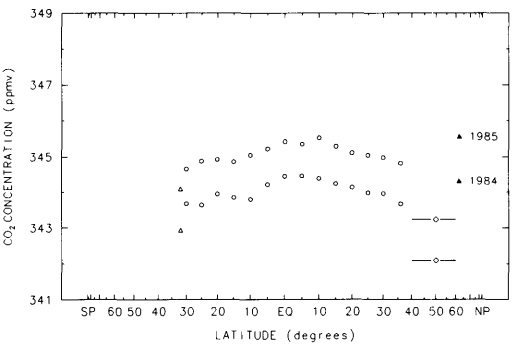

Fig. 4. Yearly mean CO_2 concentrations in the upper troposphere (open circles), at 7.2 km height over Anchorage (closed triangles) and Sydney (open triangles), and in the lower stratosphere (rectangles) in 1984 and 1985.

Table 1. *Yearly mean CO_2 concentrations at respective locations in 1984 and 1985, and standard deviations from curve fitting to the data*

Location	Altitude (km)	CO_2 (ppmv) 1984	CO_2 (ppmv) 1985	SD (ppmv)
Anchorage	7.2	344.3	345.6	0.64
36°N	↑	343.7	344.8	0.33
30°N		344.0	345.0	0.23
25°N		344.0	345.1	0.56
20°N		344.1	345.1	0.50
15°N		344.2	345.3	0.41
10°N		344.4	345.5	0.31
5°N		344.5	345.4	0.23
0°	10 ~ 12	344.5	345.4	0.42
5°S	↓	344.2	345.2	0.32
10°S		343.8	345.1	0.23
15°S		343.9	344.9	0.23
20°S		344.0	344.9	0.22
25°S		343.6	344.9	0.22
30°S		343.7	344.7	0.21
Sydney	7.2	342.9	344.1	0.23
40°–60°N	lower stratosphere	342.1	343.2	0.13

Average values of yearly mean CO_2 concentrations in 1984 and 1985 are compared in Fig. 5 with those in the lower troposphere. The lower tropospheric values were obtained from (1) our ship-board measurements between Yokohama, Japan and Melbourne, Australia (Tanaka et al., 1987b) and between Tokyo, Japan and Seattle, USA (unpublished data), (2) aircraft measurements in the height interval of 0–1 km above the Pacific Ocean off the coast of Sendai (Tanaka et al., 1987a), and (3) continuous measurements at Syowa Station, Antarctica (Tanaka et al., 1987c; Nakazawa et al., 1991). Typical sampling routes of ship-board measurements and locations of Sendai (38°N) and Syowa Station (69°S) are shown in Fig. 1. The results of continuous measurements at two GMCC stations of Pt. Barrow, Alaska (71°N)

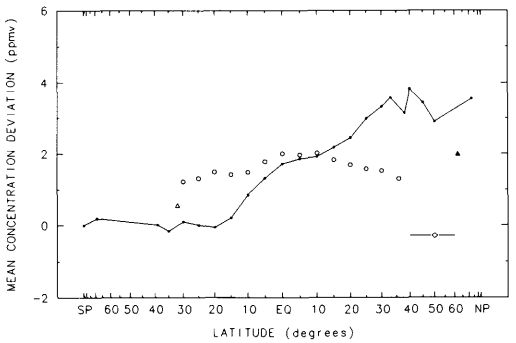

Fig. 5. Average values of yearly mean CO_2 concentrations of 1984 and 1985 in the lower (closed circles with solid line) and upper (open circles) troposphere, at 7.2 km height over Anchorage (closed triangle) and Sydney (open triangle), and in the lower stratosphere (rectangle). Values are represented as a deviation from the South Pole.

and South Pole (Schnell, 1986) are also plotted in Fig. 5 for reference. Measurement systems used in GMCC and Tohoku University have been developed independently. Nevertheless, continuous measurements at South Pole and Syowa from 1984 to 1987 showed that yearly mean CO_2 concentrations at both stations agree well with each other, values of Syowa being higher by only 0.1 ppmv, on average, than those of South Pole (Nakazawa et al., 1991). The concentration of atmospheric CO_2 can vary longitudinally (Tans et al., 1989), but the general feature of the global CO_2 distribution can be seen in Fig. 5. In the northern hemisphere, lower tropospheric values are higher than upper tropospheric values, and the situation is reversed in the southern hemisphere. Vertical differences in CO_2 concentration amount to about 2.0 ppmv in high and middle latitudes of the northern hemisphere, reflecting the fact that a large quantity of CO_2 is released into the atmosphere by fossil fuel combustion (Rotty, 1983). This difference agrees well with those from our aircraft measurements over Japan (Tanaka et al., 1983a, 1987a, 1988). Such differences disappear in northern low latitudes and extend inversely southward until values of about 1.3 ppmv are observed south of 15°S. The vertical CO_2 profile in the southern hemisphere is attributed to southward transport of the northern hemispheric air through the upper troposphere, to be discussed later, and to the CO_2 uptake by the southern hemispheric oceans (Tans et al., 1989).

Fig. 6 shows the average seasonal cycle of the CO_2 concentration approximated by eq. (1). The seasonal CO_2 cycle in the upper part of the troposphere is most enhanced in northern high latitudes and decreases gradually, going southward with a phase delay, but is still discernable in the southern hemisphere where the seasonal cycle of the lower tropospheric CO_2 is very small (Komhyr et al., 1985; Beardsmore and Pearman, 1987; Tanaka et al., 1987b, c; Conway et al., 1988; Nakazawa et al., 1991). The seasonal CO_2 cycle at 7.2 km height over Anchorage shows maximum and minimum concentrations in late April and in late August or early September, respectively, with a peak-to-peak amplitude of 9.4 ppmv. The phase agrees well with the result of aircraft measurements made over northern high latitudes from 1963 to

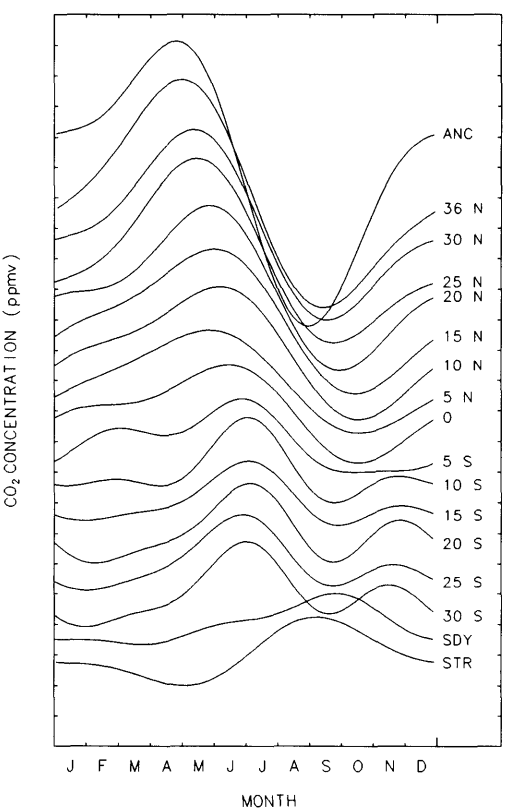

Fig. 6. Seasonal CO_2 cycles in the upper troposphere at respective latitudes, at 7.2 km height over Anchorage (ANC) and Sydney (SDY), and in the lower stratosphere (STR). One division of the ordinate corresponds to 1 ppmv.

1968 (Bolin and Bischof, 1970), but the amplitude observed here is larger by almost 1.0 ppmv. This difference in amplitude may be due to enhanced metabolic activity of the land plants. Such an increase of the amplitude was also found at various places around the world (Pearman and Hyson, 1980; Bacastow et al., 1981, 1985; Cleveland et al., 1983; Keeling et al., 1985).

The concentration of the upper tropospheric CO_2 over Japan reaches a maximum late in April or early in May and a minimum in mid-September, and its peak-to-peak amplitude is 7.6 ppmv. The seasonal CO_2 cycle is delayed with decreasing latitude and, at the equator, maximum and minimum concentrations are found in mid-June and in mid-October, respectively. The amplitude also decreases going southward until nearly constant values of about 2.5 ppmv are observed in the southern hemisphere.

The character of the seasonal CO_2 cycle is changed south of 10°S; high and low concentrations are found twice a year. Such variations could be explained by suppression and enhancement of southward transport of the northern hemispheric air associated with the South Pacific Convergence Zone (SPCZ) and the monsoon circulation. The SPCZ is usually located between 0° and 10°S on our flight routes from December to April. The lower tropospheric air in both hemispheres converges there, ascends and then returns to each hemisphere through the upper troposphere. Therefore, an inter-hemispheric air exchange is largely suppressed and the CO_2 concentrations are changed discontinuously, as seen in Fig. 7. Such a phenomenon was also found in the lower troposphere (Tanaka et al., 1987b). During this time of the year, the upper tropospheric CO_2 in the southern hemisphere shows low concentrations due to the mixing of its own hemispheric air. Slow increase of the concentration for this period may be due to horizontal diffusion of the northern hemispheric air through the SPCZ. In this connection, the northern hemispheric air is richer in CO_2 than the southern hemispheric air during this period except in December, and the concentration difference between both hemispheres increases gradually with time. In May, when the difference reaches a maximum, the monsoon circulation begins to occur and the SPCZ disappears, and the northern hemispheric air begins to intrude into the southern hemisphere. Since this intrusion continues until November, the CO_2 concentration reaches a maximum late in June or early in July, and a minimum late in September, and then increases again.

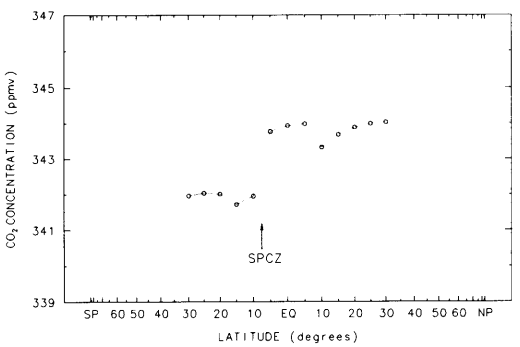

Fig. 7. Latitudinal distribution of the upper tropospheric CO_2 concentration between Tokyo and Sydney on February 22, 1984.

Seasonal CO_2 cycles with almost the same amplitude and phase are observed in the upper troposphere south of 10°S. However, the seasonal CO_2 cycle at 7.2 km height over Sydney is quite different from those; maximum and minimum concentrations occur late in September and late in March, respectively, and the peak-to-peak amplitude is 1.7 ppmv.

From the above, it is suggested that southward transport of the northern hemispheric air by the monsoon circulation occurs very rapidly only through the upper troposphere, and the transported air is mixed slowly with the southern hemispheric air. The air transport from the northern hemisphere to the southern hemisphere through the upper troposphere was also suggested by Nikaidou (1989) and Pearman and Beardsmore (1984). Nikaidou showed, from numerical experiments on the transport of tracer particles, that this transport process is much enhanced in the summer season of the northern hemisphere. Pearman and Beardsmore found from their aircraft measurements over Australia that the seasonal CO_2 cycle is larger in the upper troposphere than in the middle troposphere and that the phase precedes in the upper levels of the troposphere.

The seasonal CO_2 cycle observed by Pearman and Beardsmore in the middle troposphere shows the minimum concentration in April and the maxi-

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mum concentration in October, and peak-to-peak amplitude of 1.2 ppmv. This cycle is similar to ours at 7.2 km height over Sydney, but the amplitude is smaller by 0.5 ppmv. In their upper troposphere results, low values of the CO_2 concentration are seen between November or December and April or May, and high values during the remaining period of the year. A comparison of their results with ours, shows similar periods with low CO_2 concentrations but the onset of high concentration is delayed and lasts longer in their results. This may be due to different areas and years in the air sampling between the 2 sets of measurements. Pearman and Beardsmore also found that the seasonal cycle of the upper tropospheric CO_2 is enhanced in lower latitudes. It may be suggested from this fact that the influence of the air transported from the northern hemisphere in association with the monsoon circulation is different for different latitudes.

The peak-to-peak amplitudes of the seasonal CO_2 cycle in the upper and lower troposphere are shown in Fig. 8. The seasonal CO_2 cycle observed in these 2 sets shows similar latitudinal characteristics; they are enhanced in northern high latitudes and decrease southward. However, inspection indicates qualitatively different facets. The amplitudes at 7.2 km height over Anchorage and in the upper troposphere over Japan are almost half of the lower tropospheric values. The vertical difference of the amplitude is largely reduced near 30° N, diminishes gradually with decreasing latitude and is reversed in the southern hemisphere, at least in the region covered by this measurement. Fig. 9 shows the occurrence of maximum and minimum concentrations of the seasonal CO_2 cycle in the upper and lower troposphere of the northern hemisphere. The CO_2 concentration at 7.2 km height over Anchorage varies seasonally in phase with that of the lower tropospheric air. Over Japan, the seasonal cycle of the lower tropospheric CO_2 is advanced by about 1 month more than that of the upper tropospheric CO_2, reflecting the fact that our lower tropospheric data were taken in proximity to Japan and the Asian Continent. The altitude-dependent phase shift is also seen in the maximum concentration in low latitudes. The minimum concentration occurs in mid-October for both the upper and lower troposphere, due to northward transport of the southern lower tropospheric air by the monsoon circulation (Tanaka et al., 1987b).

In the southern hemisphere, the seasonal CO_2 cycle is clearly observable in the upper troposphere as a result of the air transport from the northern hemisphere, but it is extremely small in the lower troposphere due to the suppression of southward transport of the northern lower tropospheric air by

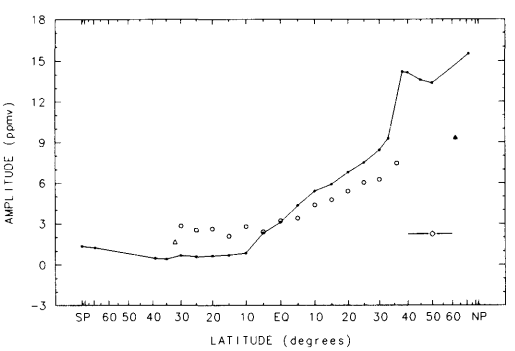

Fig. 8. Peak-to-peak amplitudes of the average seasonal CO_2 cycle in the lower (closed circles with solide line) and upper (open circles) troposphere, at 7.2 km height over Anchorage (closed triangle) and Sydney (open triangle), and in the lower stratosphere (rectangle).

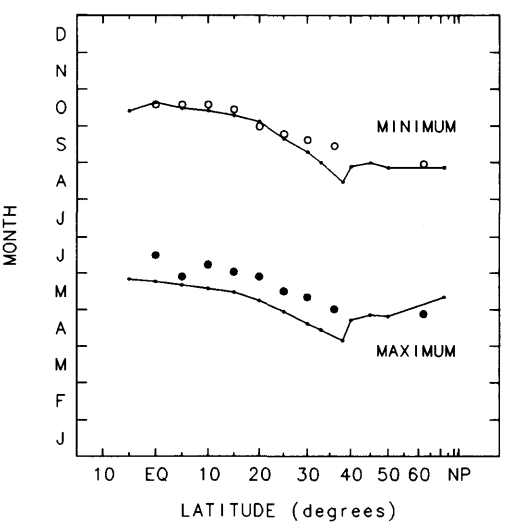

Fig. 9. Time of occurrence of the maximum and minimum concentrations of the average seasonal CO_2 cycle in the lower (closed circles with solid lines) and upper (closed and open circles) troposphere in the northern hemisphere.

the ITCZ and SPCZ, no seasonality in low latitudes and the smaller area of austral vegetated lands. This measurement was made as far as 30° S. It is interesting to know the temporal and spatial behavior of the upper tropospheric CO_2 concentration in regions further south. This knowledge is very useful to understand the transport process of atmospheric CO_2 to the south polar region.

Yearly mean values of the lower stratospheric CO_2 concentration are the lowest values observed in this measurement program and are lower by 1.4 and 2.1 ppmv than those of the upper troposphere over Japan and at 7.2 km height over Anchorage, respectively, as seen in Fig. 4. The difference in yearly mean CO_2 concentrations between 1984 and 1985 is 1.1 ppmv, which is in good agreement with those of the upper troposphere.

As shown in Fig. 6, the seasonal cycle of the lower stratospheric CO_2 shows a minimum concentration early in May, a maximum concentration early in September, and a peak-to-peak amplitude of 2.2 ppmv. Similar cycle was also observed by Bischof (1985) over northern high latitudes. Such a seasonal CO_2 cycle is caused by air transport from the troposphere, since there are no CO_2 sources in the stratosphere. Two possibilities are considered for this transport process, i.e., vertical diffusion of the tropospheric air through the tropopause and upwelling of the tropospheric air by the cumulous convection near the ITCZ and SPCZ. However, the former may be a minor effect, because the CO_2 concentration of the lower stratospheric air varies seasonally out of phase with that of the upper tropospheric air, as seen in Fig. 6. Therefore, the latter may be more effective for the above process, as suggested by Kida (1983).

The upper tropospheric CO_2 in the equatorial region shows the seasonal cycle with the maximum concentration in mid-June, the minimum concentration in mid-October and a peak-to-peak amplitude of about 3.0 ppmv. Compared with this cycle, the maximum of the seasonal cycle of the lower stratospheric CO_2 is delayed by about 3 months and the minimum, by nearly 7 months. Such a phenomenon may be partly attributed to seasonally-dependent transport of the tropospheric air into the stratosphere, since the ITCZ and SPCZ become active in winter and spring. On the other hand, the amplitude is smaller by only 1.0 ppmv than the upper tropospheric values in the equatorial region (see Fig. 6). Even if the tropospheric air intrudes into the stratosphere in low latitudes and then reaches northern high latitudes within a few months, it is difficult to imagine it causing an amplitude as large as about 2.0 ppmv there. Therefore, some mechanism seems to exist in the lower stratosphere to extend the seasonal CO_2 cycle. Vertical mixing of the lower stratospheric air by the jet stream may be responsible for this mechanism; the jet stream becomes active in winter and spring, and the CO_2 concentration decreases with height in the lower stratosphere (Gamo et al., 1989). As a result of such a mixing, low values of the CO_2 concentration are found in these seasons. In summer, the activity of the jet stream is largely reduced and the air transported from low latitudes play an important role for variations of the lower stratospheric CO_2. The vertical mixing of the lower stratospheric air during the cold months may also be responsible for the different phase shifts for the maximum and minimum concentrations of the seasonal CO_2 cycle mentioned above. However, further studies are necessary to confirm this hypothesis. It is especially important for this purpose to observe variations of the stratospheric CO_2 in low latitudes.

5. Conclusions

To elucidate spatial and temporal variations of the upper tropospheric and lower stratospheric CO_2 over a wide geographical area, systematic collection of air samples was made between Tokyo and Anchorage, and between Tokyo and Sydney for 2 years, 1984-1985. Yearly mean values of the upper tropospheric CO_2 concentration were high in the equatorial region, due to upward transport of lower tropospheric air near the ITCZ, and decreased gradually poleward. In the northern hemisphere, yearly mean CO_2 concentrations were lower in the upper troposphere than in the lower troposphere; the situation was reversed in the southern hemisphere. The seasonal CO_2 cycle in the upper troposphere was most enhanced in northern high latitudes and decreased going southward with a phase delay, but still prominent in low and middle latitudes of the southern hemisphere where the seasonal cycle of the lower tropospheric CO_2 is barely observable. The upper

tropospheric seasonal CO_2 cycle was more complicated in the southern hemisphere than in the northern hemisphere; high and low CO_2 concentrations were found twice a year. It is suggested from these results that a large amount of CO_2 is released from the surface of middle and high latitudes of the northern hemisphere, that the released CO_2 is transported effectively to the southern hemisphere through the upper troposphere, and that this southward transport of CO_2 is much enhanced during the period from May to November when the monsoon circulation occurs in Southeast Asia.

Yearly mean values of the lower stratospheric CO_2 concentration in northern high latitudes were lower by about 1.4–2.2 ppmv than the upper tropospheric values, and the annual CO_2 increase rate of 1.1 ppmv/year agreed quite well with that of the upper troposphere. The clear seasonal cycle with peak-to-peak amplitude of 2.2 ppmv was found in the lower stratosphere. Vertical mixing of the lower stratospheric air by the jet stream as well as upwelling of the tropospheric air by the cumulous convection near the ITCZ and SPCZ appear to be important in order to interpret this seasonal CO_2 cycle.

The results obtained in this measurement are useful for developing and validating carbon cycle models from which spatial and temporal variations of CO_2 sources/sinks on the earth's surface are deduced. However, in addition to 2 programs presently operating over Japan and Australia, further aircraft measurements are required for a better understanding of the global distribution of atmospheric CO_2, especially in areas without substantial influence of the monsoon circulation, in the equatorial and polar regions.

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