Tellus (1995), 47B, 375–390
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TELLUS
ISSN 0280–6509

Concentration variations of atmospheric CO_2 over Syowa Station, Antarctica and their interpretation

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(Manuscript received 18 April 1994; in final form 21 March 1995)

ABSTRACT

Aircraft and ground-based measurements of the atmospheric CO_2 concentration have been made at Syowa Station, Antarctica since January 1983. The minimum concentration of the average seasonal CO_2 cycle occurs in March throughout the troposphere, while the maximum concentration appears in mid-August in the upper troposphere and in late September in the lower and middle troposphere. The peak-to-peak amplitude of the cycle decreases slightly with height. The CO_2 concentration increases with height during most of the year, but from late winter to spring this height dependency is minimal. To examine the contribution of the atmospheric transport processes to these variations of the CO_2 concentration, a 3-dimensional trajectory analysis was performed using data from the US National Meteorological Center. From the results obtained, it is postulated that northern hemispheric air with relatively high CO_2 concentration is transported to the antarctic region through the upper troposphere from late fall to winter, while air with low CO_2 concentration is transported from the southern hemisphere middle latitudes into the antarctic region through the lower troposphere in the remaining seasons. It is hypothesized that these air transport processes could influence CO_2 variations over the station.

1. Introduction

Systematic measurements of the background concentration of atmospheric CO_2 have been recently made at many sites around the world (e.g., Conway et al., 1991; Keeling, 1991). The data from these measurements are necessary to elucidate the carbon cycle on the earth's surface, but further measurements are required for a better understanding of the cycle (Pearman and Hyson, 1986; Fung et al., 1983; Heimann et al., 1989; Keeling et al., 1989a, b; Tans et al., 1989, 1990; Enting and Mansbridge, 1991).

Temporal and spatial variations of atmospheric CO_2 at different sites are influenced by many factors, e.g., anthropogenic emissions (fossil fuel combustion and deforestation), air-sea exchange, air-land biosphere exchange, and atmospheric transport processes. The CO_2 variations are thought to be largely affected by air transport processes from different locations, especially in regions isolated from natural sources/sinks and anthropogenic sources. Since there are no strong CO_2 sources and sinks in the antarctic region, it is likely that systematic measurements of the CO_2

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concentration in this region will give us not only detailed information on temporal variations of the atmospheric CO_2 concentration, such as the secular trend and its small year-to-year fluctuations (Nakazawa et al., 1991a), but also information on the long-range atmospheric transport processes of CO_2.

Systematic measurements of CO_2 concentration in the antarctic region were taken for the first time by C. D. Keeling at the South Pole (SPO) in 1957 (Keeling et al., 1976); now 5 ground-based stations are operating in the antarctic region (Beardsmore and Pearman, 1987; Conway et al., 1988; Nakazawa et al., 1991a). We initiated CO_2 flask sampling at Syowa Station (SYO; 69°00′S, 39°35′E) in January 1983 and continuous measurement in February 1984 (Tanaka et al., 1987a).

Nakazawa et al. (1991a) suggested that CO_2 variations in the antarctic region are caused not only by oceanic and biospheric CO_2 exchange in the southern hemisphere but also by air transport processes. However, the sparsity of surface stations and the smallness of variations at southern mid to high latitudes have made clear delineation of these processes difficult (Pearman and Hyson, 1986; Conway et al., 1988). For a better understanding of CO_2 variations in the antarctic region in terms of atmospheric transport processes, we have continued, in addition to the ground-based measurements, aircraft measurements of atmospheric CO_2 concentration over SYO since January 1983 (except in 1985 and 1988). We have also performed a 3-dimensional trajectory analysis using data from the US National Meteorological Center (NMC) and compared its result with meridional CO_2 concentration field constructed from the observed data. In this paper, the results of the aircraft measurements and the trajectory analysis are presented, and variations of CO_2 concentration in the antarctic region are discussed.

2. Aircraft measurements

Except for 1985 and 1988, aircraft measurements have been made frequently each year over SYO since January 1983 during times outside of polar nights and bad conditions of the airfield constructed on fast ice. SYO is located on East Ongul Island (69°00′S, 39°35′E) in Lützow-Holm Bay, away from the Antarctic Continent by about 4 km. Detailed descriptions of SYO are given by Nakazawa et al. (1991a) and Tanaka et al. (1987a). Measurements were made once or twice in a month when flight operations were made. The aircraft used for collecting air samples was a Pilatus Poter PC-6. The air samples were obtained at the 300 m level, and about every kilometer between 1-km and approximately 7-km height levels; the samples were taken from the wingtip to avoid contamination from the engine exhaust. The sampled air was filled into 550 ml Pyrex glass flasks at an over-pressure of 2.0×10^5 Pa using an electric diaphragm pump. Before air sampling, each flask was washed sufficiently in an ultra-sonic bath with purified water, dried, and then was evacuated to lower than 0.13 Pa at about 100°C for at least 5 hours.

The air samples collected were returned to our laboratory and their CO_2 concentrations were determined by a non-dispersive infrared analyzer with a precision of ± 0.01 ppmv against 3 or 4 air-based working standard gases (Nakazawa et al., 1992). Water vapor in air samples was removed using 2 glass traps cooled at -78°C. The concentrations of the working standard gases were calibrated with the above-mentioned non-dispersive infrared analyzer against our secondary standard gases before and after their use. The secondary standard gases were calibrated once or twice a year against our primary standard gases whose concentrations were determined gravimetrically with an absolute accuracy of ± 0.13 ppmv. The concentration drifts of the working standard gases used for this measurement were less than 0.05 ppmv. Because of the time gap (t) (420 days in some cases) between the time of actual air sampling and subsequent laboratory analysis, a correction was necessary to compensate for the possible deterioration of the air samples during their storage in flasks. For the data after February 1984, the degree of deterioration was estimated from the concentration differences (ΔC) between the continuous in-situ measurements and flask samples at each flask sampling time of about once a week at SYO. Next, ΔC data were fitted to a polynomial function of t. The fitting functions were calculated for the data of each year because year-to-year differences in the distribution of ΔC were found. Since the continuous CO_2 measurements started on February, 1984, the data before and including January, 1984 were fitted with functions

estimated from laboratory experiments where dry standard gases were filled in the sample flasks for a long time (see Fig. 3 in Tanaka et al. (1987a)). The standard deviation of the data from each fitting function was 0.28 ppmv on average. Finally, these fitting functions were applied to the aircraft data and then the corrected concentrations were estimated. A maximum correction obtained from this procedure amounted to 3.1 ppmv.

3. Trajectory analysis

Details of the trajectory analysis are described by Yamazaki et al. (1989) and Yamazaki (1992). Therefore, only a brief description and supplemental explanations are presented here. The 3-D twice-daily US NMC meteorological data were used in the trajectory analysis. Its horizontal resolutions were 2.5° by 2.5° in latitude and longitude, with data at 12 standard levels between 1000 hPa and 50 hPa. Vertical wind velocities were obtained from horizontal wind field using a mass continuity equation under the assumption of $\omega = 0$ at 30 hPa. The time step for the calculation was set to 1 hour, and 3-D wind at the position of an air parcel at each time step was obtained from the winds at the surrounding grid points by temporal and spatial linear interpolation.

Trajectory models are typically integrated for 5 days in order to minimize the violation of some of the basic assumptions in the tracing technique, such as constant acceleration and no mixing of air parcels. Trajectory analysis is very sensitive to errors in the meteorological data used to calculate the paths of air parcels. Regions of divergent flow, for example, can cause almost chaotic dispersion of air parcels, so that air within a certain small region may appear to originate from many diversely different regions in a backward trajectory calculation. Vertical velocity calculation based on a mass continuity equation is also very sensitive to meteorological data since it is the result of a difference between two large terms (Holton, 1979). It is interesting to note however that most trajectories calculated in this fashion follow isentropes (Yamazaki et al., 1989) which take into account the vertical motion of the air parcels, and that away from any significant diabatic influences (such as synoptic storms) isentropic surfaces are good approximation to material surfaces (Danielsen, 1961). Thus, for the trajectory analysis to be of any use identifying certain source-receptor relationships (in this case, between the atmospheric CO_2 concentration field and the CO_2 variation observed at SYO), "details of individual trajectories should be ignored in favor of indications of the general directions of motion gained from swarms of trajectories from nearby points." (Merrill, 1989). It is within this statistical context that we cautiously interpret the results of the trajectory analysis. The relationships derived from the trajectory analysis between the atmospheric CO_2 concentration field and the CO_2 variation over SYO are viewed as highly qualitative and suggestive, and certainly not conclusive.

The backward trajectory calculations were performed for 30 days because the time scale of meridional transport and mixing is on the order of a month, while the zonal mixing is typically accomplished within a week or so. There is generally more inhomogeneity in the atmospheric CO_2 in the meridional direction than in the zonal direction. In addition, 30-day back trajectories give statistically more distinctive or resolvable distribution of the trajectory "origins" than, say 10 day back trajectories, although admittedly certain percentage of the distinctiveness in the distribution could be attributable to the violation of the assumptions, as well as to all the uncertainties associated with the trajectory calculations. Thus, within these limitations, we cautiously employ trajectory analysis to examine if we could obtain physically reasonable and consistent interpretation of the CO_2 variation observed at SYO in terms of seasonal transport of air parcels with relatively high and low CO_2 concentrations to SYO. We will show in the next section that trajectory results are, in general, consistent with suggestions made by Nakazawa et al. (1991a).

The following analysis procedures were performed: (1) Atmospheric transport processes at every 50 hPa between 350 and 850 hPa in pressure level over SYO were examined. For this purpose, 143 (11×13) air parcels were initially distributed at the grid points with the interval of 0.4° in longitude and 0.2° in latitude at each pressure level over SYO (Fig. 1). (2) Backward trajectories of these parcels for 30 days were calculated for each pressure level to obtain the distributions of the parcels 30 days before. The arrival time at each level over SYO was set at 12Z every 2 days from 1 January to 31 December 1989. Trenberth and

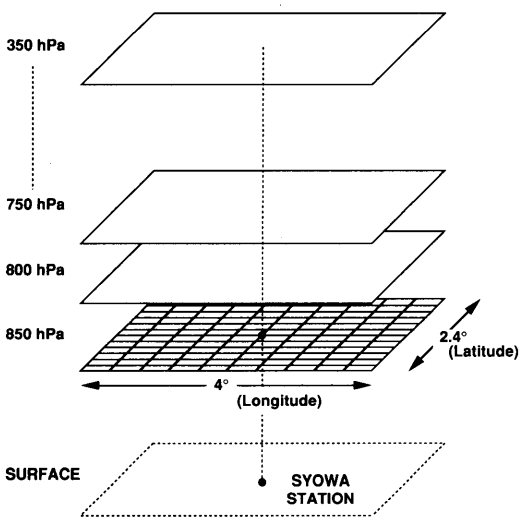

Fig. 1. Schematic illustration of the initial distribution of air parcels for trajectory analysis.

Olson (1988) have shown that the NMC analyses in the antarctic region prior to May 1986 are unreliable. (3) 15 or 16 calculated distributions of the parcels which arrive at each selected level over SYO each month were superimposed on one another. (4) From this resultant distribution of air parcels, the number of parcels in each meridional box with 100 hPa and 10° intervals were counted. The same procedures were applied to all selected levels and months during the above-mentioned period of study. The height-latitude distributions of the air parcels were used for interpreting the seasonal CO_2 cycles over SYO, which will be discussed later.

4. Results and discussion

4.1. Aircraft measurements

Aircraft measurements of the CO_2 concentration were first grouped into 3 height intervals of <2.5, 2.5–5, and >5 km. The data classified into these respective height intervals were then simply averaged. Furthermore, we only consider the average seasonal cycle and its height dependency in this paper. To resolve the secular increase and seasonal cycle in the CO_2 measurement, a digital filtering technique, which includes Fourier harmonics, Reinsch-type spline and Butterworth filter, was used (Nakazawa et al., 1991b, 1992). Using this technique, the following analysis procedures were performed.

(1) *Removal of secular trend from data at each height interval.* Since it was difficult to estimate the secular increase at each height interval, the secular trend calculated for the surface-based data at SYO was assumed to be applicable at each interval. That this procedure provides a good approximation of the secular trend at all heights is based on the height-independent correlation of secular variations of CO_2 throughout the troposphere over Japan (Nakazawa et al., 1993a; Tanaka et al., 1987b).

(2) *Determination of average seasonal cycle at each height interval.* Composite analysis was performed for each height interval by superimposing detrended 1-year data set on top of each other, and then fitting the fundamental and its first harmonics. Outliers were then removed and the fitting procedure was repeated to obtain average seasonal cycle for the period of record for each of the height

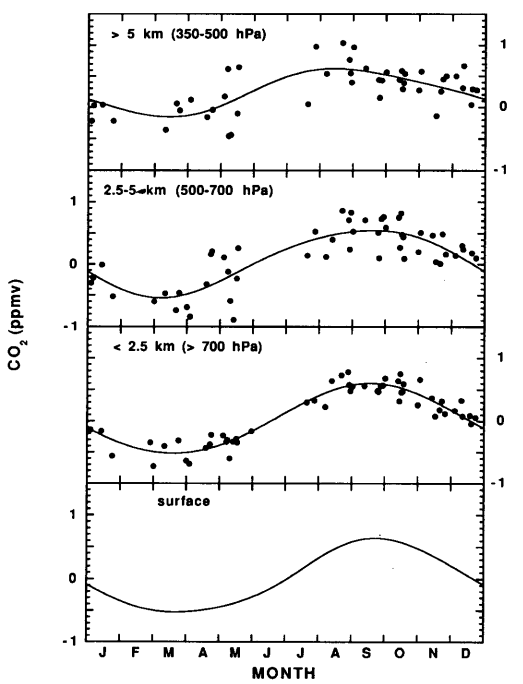

Fig. 2. Average seasonal CO_2 cycles over Syowa Station. Respective values are shown as deviations from the mean value at the surface during January 1983–January 1992.

intervals. The numbers of rejected data were 2 out of 56, 3 out of 55 and 3 out of 49, for the <2.5, 2.5–5, >5.0 km intervals, respectively.

Average seasonal CO_2 cycle at each height interval is shown in Fig. 2, relative to the mean surface value for the period from January 1983 to January 1992. Average seasonal cycle at the surface is also shown for reference. Several plotted data appear scattered. This is attributable to the imperfect correction for sample deterioration. Standard deviations of the data from the fitted seasonal curves are 0.12, 0.13, 0.26 and 0.27 ppmv for the surface, <2.5, 2.5–5, >5 km height intervals, respectively. The minimum CO_2 concentration occurs in March at all heights, while the maximum appears in mid-August in the >5.0 km layer and in late September at the surface and in the <2.5 and 2.5–5.0 km layers. The peak-to-peak amplitudes are 1.2, 1.1, 1.1 and 0.8 ppmv for the surface, <2.5, 2.5–5, >5 km layers, respectively, showing a slight decrease with height. The results of the 3-D transport modeling studies by Heimann et al. (1989) and Fung et al. (1987) are consistent with the above observations.

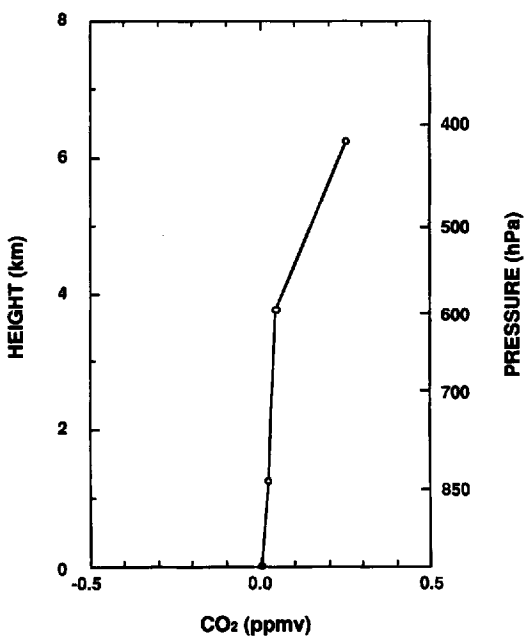

Fig. 3. Annual mean vertical profile of the CO_2 concentration over Syowa Station. Respective values are shown as deviations from the mean value at the surface during January 1983–January 1992.

The CO_2 concentration increases slightly with height during most of the year, but from late winter to spring this height dependency is minimal. A vertical distribution of the mean CO_2 concentration for the period covered by the study is shown in Fig. 3. The surface-to-top difference is about 0.3 ppmv. Similar vertical profiles have been documented at southern hemisphere (SH) middle latitudes by Pearman and Beardsmore (1984) and Nakazawa et al. (1991b).

There are interesting differences between the seasonal cycle observed at SYO and those reported by Nakazawa et al. (1991b) for the SH middle latitude region, and Beardsmore and Pearman (1987, 1991a, b) for Cape Grim, Tasmania (CGO).

(1) *In contrast to SYO, the seasonal cycle in the SH midlatitude upper troposphere is characterized by a 2-cycle sinusoid (2 minima and 2 maxima).* Greater of the 2 maxima in the seasonal cycle in the upper troposphere at 30°S occurs in early July, about 1.5 months earlier than the maximum observed in the >5.0 km layer over SYO. On the other hand, the southern midlatitude seasonal cycle in the low to mid troposphere is similar to the one observed over SYO for the corresponding tropospheric layers, with maximum occurring around the same time for both regions.

(2) *In contrast to SYO, the peak-to-peak amplitude in the seasonal cycle at SH midlatitude region increases dramatically with height.* The amplitude in the upper troposphere at 30°S is more than 3 times as large as that seen in the >5 km layer over SYO, though the amplitude at CGO is slightly smaller than observed at SYO (Nakazawa et al., 1991a).

(3) *Unlike at SYO, the increase with height of CO_2 concentration during much of the year in the SH midlatitudes is much more pronounced.*

(4) *CO_2 concentration is higher at SYO than at CGO throughout the year.* However, the concentration difference is larger in spring than in late summer to early winter (Nakazawa et al., 1991a; Beardsmore and Pearman, 1991a, b).

Both Pearman and Beardsmore (1984) and Nakazawa et al. (1991b) have suggested that the transport processes of bringing air with relatively high CO_2 concentration into the southern hemisphere from the northern hemisphere through the upper troposphere play an important role in causing variations of the upper tropospheric CO_2

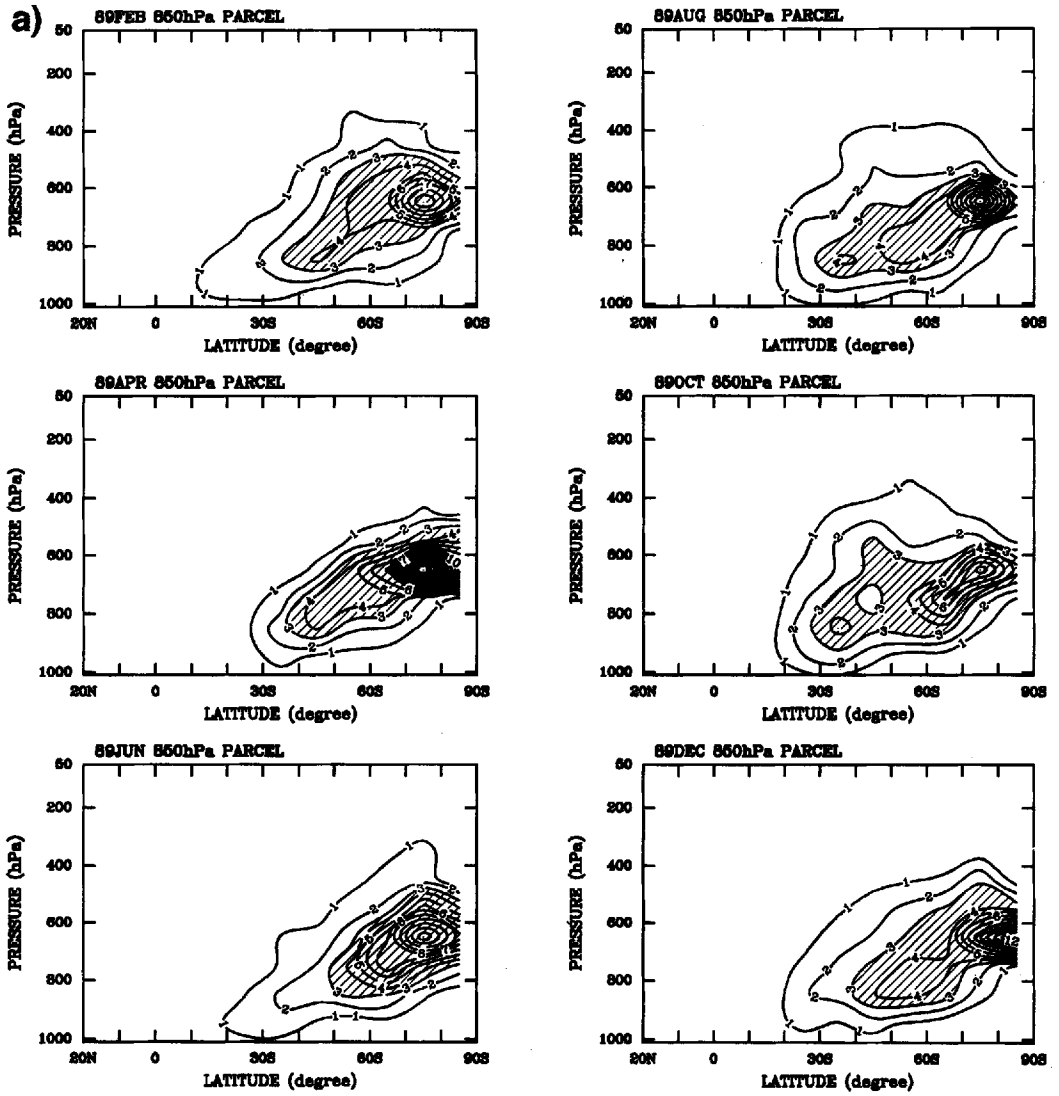

Fig. 4. Height-latitude distributions of the air parcels 30 days before being transported to 850 hPa (a) and 400 hPa (b) over Syowa Station in even months of 1989. Each month in this figure refers to the beginning of the back trajectory. Values are represented as the percentage ratio of the number of air parcels included in each meridional box with 100 hPa and 10° intervals to the total number of the parcels transported in the month. The areas being more than 3% are shaded.

at SH low and middle latitudes. It is quite possible then that the CO_2 measurements over SYO are influenced by the same northern hemisphere air, particularly in the upper troposphere. But the fact that the seasonal CO_2 cycle at SYO decreases in amplitude with height seems to indicate there are other transport processes which could be important for interpreting the CO_2 variations in the antarctic region. This will now be discussed.

4.2. *Trajectory analysis*

It should be mentioned from the outset that the results in this section, although presented and discussed in somewhat definitive manner, should be

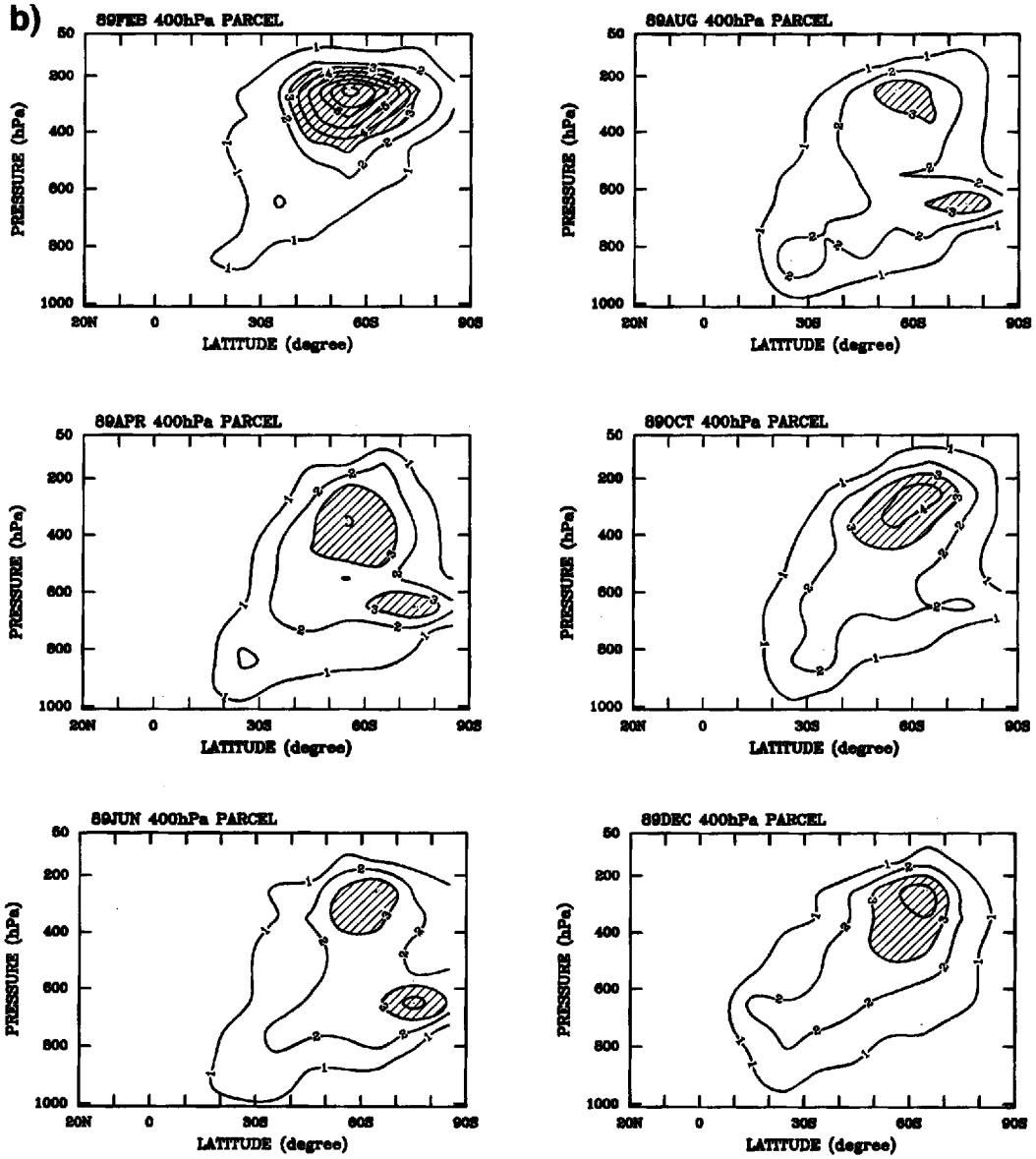

Fig. 4 (cont'd).

viewed with all the caveats mentioned in Section 3. However, the results are in general consistent with the speculations made by Nakazawa et al. (1991b), Pearman and Beardsmore (1984) and Pearman and Hyson (1986).

Figure 4 shows height-latitude distributions of the trajectory "origins" of the air parcels 30 days before being transported to 850 hPa and 400 hPa over SYO for the even months in 1989. Each month in this figure therefore refers to the beginning of the back trajectory. Values represent percentage ratios of the number of air parcel "origins" in individual boxes to the total number of air parcels. This figure shows that the air parcels

transported to 850 hPa level over SYO originate mainly in the middle troposphere over Antarctica and partly in the lower troposphere at lower latitudes. Most of the air parcels are transported adiabatically (Yamazaki et al., 1989). Such distribution of the air parcels is so dominant throughout the year that its seasonal variation is small. On the other hand, in the case of 400 hPa (Fig. 4b), the air parcel distributions are more dispersed and cover a wider area compared to those for 850 hPa. Though many parcels are transported from the upper troposphere at lower latitudes (about 60°S) throughout the year, a considerable number of the parcels are also transported from the lower levels at SH high latitudes from late fall to winter. The latter transport is largely suppressed in summer. The cyclone activity at the circumpolar trough zone is enhanced in cold seasons and suppressed in summer. This seasonal variation of cyclone activity is probably associated with the seasonal variation of atmospheric transport.

In the following, we obtain estimates of the contribution of atmospheric transports originating from different locations in the temporally varying atmospheric CO_2 concentration field to the observed CO_2 variations at different heights over SYO. The analysis was performed as a modeling exercise, and seasonal variations of CO_2 over SYO are discussed on the basis of its results.

The height-latitude distribution of monthly mean CO_2 relative to the SYO mean for the period January 1983 to January 1992 constructed from the observed data was used for this analysis. Distribution fields for the odd months are shown in Fig. 5. Details of the data and the method used to derive the concentration fields are given in the Appendix. Although Fig. 5 shows height-latitude profiles of the CO_2 concentration fields, the data used for the construction of these profiles were not zonally averaged. For example, many data values used to construct the profiles originated from the SH low to midlatitudes in the Western Pacific Ocean where upper troposphere is influenced by the northern hemispheric air brought in by the monsoon circulation (Nakazawa et al., 1991b; Taguchi, 1994). This could lead to an overestimation of the importance of the transport of the upper tropospheric CO_2 in the SH low latitude region to the CO_2 variation at SYO. Even though there are observations showing east-west or longitudinal variation in the atmospheric CO_2 (Tans et al., 1989; Nakazawa et al., 1992) and year-to-year variation of the seasonal cycle at various monitoring stations (Komhyr et al., 1985; Conway et al., 1988; Nakazawa et al., 1991a), we assume, based on the difference between north-south and east-west atmospheric mixing time scales, that the variation of atmospheric CO_2 is on average greater in the latitudinal than in longitudinal direction. Therefore, we assume that the general zonally averaged features of the concentration fields are represented climatologically in Fig. 5.

We have also assumed that each air parcel arrives at SYO from its 30-day "origin" without mixing with the surrounding air. Back trajectories of less than 10 days or so do not delineate the transport sources of CO_2 variations over SYO. Therefore, the period of the trajectory analysis is extended to 30 days, though non-mixing assumption may not be completely realized. Furthermore, influence on the air parcel CO_2 concentration from sources and sinks along its trajectory path is not considered. Since the amount of air-land biosphere CO_2 exchange is small at the SH middle and high latitudes, its influence during the transport of air parcels is thought to be very small. However, within the context of the present study, the impact of air-sea exchange around Antarctica on the CO_2 concentration of the air parcels reaching SYO is very difficult to estimate. Nevertheless, we tried to come up with an order of magnitude estimate of the impact. According to Tans et al. (1990), when wind speed (W) is stronger than 3 m/s, net CO_2 flux (F) across the air-sea interface is given as follows:

$$F(\text{moles } CO_2/m^2/\text{year}) = 0.016[\,W(\text{m/s}) - 3\,] \times \Delta pCO_2(\mu\text{atm}), \quad (1)$$

where ΔpCO_2 is the pCO_2 difference between the surface ocean and the atmosphere. Now, if $\Delta pCO_2 = -17(\mu\text{atm})$ which is the annual mean ΔpCO_2 averaged in the Southern Ocean ($>50°$S) estimated by Tans et al. (1990) and $W = 10$(m/s), then $F = -1.9$(mole CO_2/m^2/year) (i.e., -5.2×10^{-3}(mole CO_2/m^2/day)). Now, atmospheric planetary boundary layer (PBL) is about 1000 m. This atmospheric layer contains roughly 45000 moles/m^2 of air. Therefore, if an air parcel travels in PBL for a whole day with a speed of 10 m/s over an area with homogeneous ΔpCO_2 of

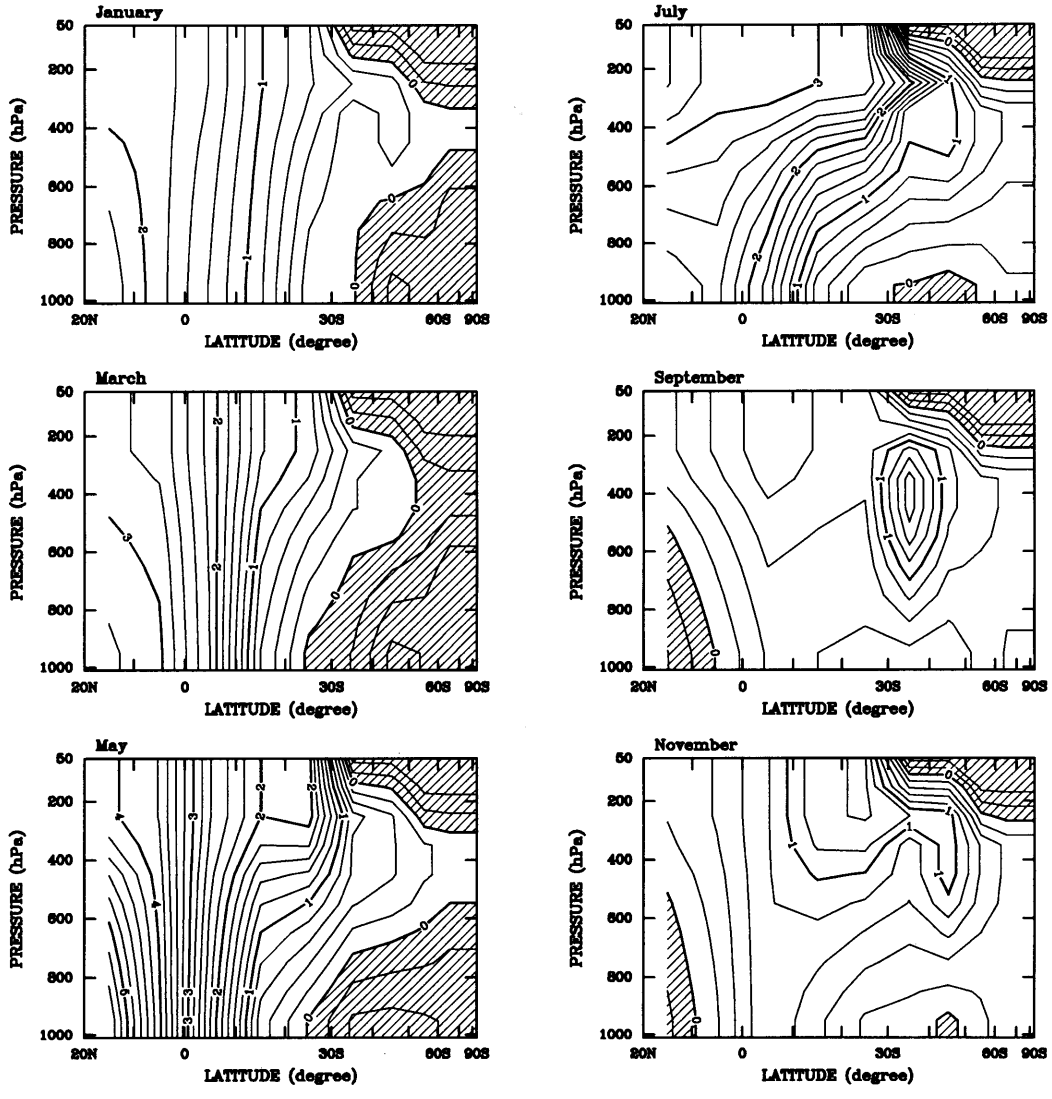

Fig. 5. Height-latitude distributions of the CO_2 concentration (ppmv) in odd months of the year constructed from observed data. Respective values are shown as deviations from the mean value at Syowa Station during January 1983–January 1992. Negative areas are shaded.

$-17\,\mu$atm, its CO_2 concentration will change roughly by -0.1 ppmv (i.e., -5.2×10^{-3}(mole $CO_2/m^2/day)/45000(moles/m^2)$). Although this is in the same ball park magnitude as the month-to-month change in the CO_2 concentration at SYO, we feel that this value represents a maximum impact on the air parcel CO_2 concentration. Surface winds are quite variable, with 10 m/s being on a high side. Furthermore, ΔpCO_2 distribution around Antarctica is very likely highly inhomogeneous (Inoue and Sugimura, 1988), with areas where $\Delta pCO_2 \sim 0$. Temporal and spatial measurements of atmospheric and oceanic CO_2 in the circumpolar region around Antarctica are very sparse and it is not fully known whether the ocean in this region acts as a net source or sink of atmospheric CO_2 (Tans et al., 1990). In summary, we do not know exactly the magnitude of the

impact of air-sea exchange might have on the CO_2 concentration of an air parcel on its route to SYO, but it is our estimation that it is relatively minimal.

It is still interesting and useful to obtain a qualitative understanding of the possible influence the atmospheric transport processes have on the observed variations of CO_2 concentration over SYO. Even though there are severe limitations to the employment of the 30-day back trajectories, such trajectory analysis does provide at least an exploratory glimpse toward that understanding. We will also show that the interpretation derived from the trajectory analysis is consistent and supports speculation put forward by earlier studies, such as Nakazawa et al. (1991a, b), Pearman and Beardsmore (1984) and Pearman and Hyson (1986). For this purpose, the distribution for the (n)th month of the year in Fig. 5 was compared with that for the $(n+1)$th month of 1989 in Fig. 4. The following are suggested from this comparison:

(1) For the lower troposphere (850 hPa), the air with relatively high CO_2 concentration is likely to be transported to SYO from around antarctic mid and upper troposphere as well as from SH low latitude troposphere. During fall and winter seasons, the concentration difference between these high concentration regions and the 850 hPa over SYO is increased. As the seasonal variation in the atmospheric transport is small, it is suggested that the high CO_2 air is transported to the lower troposphere over SYO more effectively in these seasons than in the remaining seasons; this transport coincides with the seasonally CO_2 increase observed at the station. On the other hand, air with low concentration is transported from the lower troposphere in the SH midlatitudes. During spring and summer seasons, since the concentration difference between SYO and this region is slightly increased and the high CO_2 transport from around antarctic mid and upper troposphere as well as from SH low latitude troposphere is relatively suppressed, the concentration at SYO is decreased by this low CO_2 transport. This transport process is consistent with the fact that the concentration difference between the antarctic region and the SH midlatitudes is gradually reduced from late spring to early fall (Nakazawa et al., 1991a; Beardsmore and Pearman, 1991a, b).

(2) For the upper troposphere, it is likely that air with high CO_2 concentration is transported to SYO from the SH midlatitude upper troposphere and/or from the SH low latitude troposphere throughout the year. The concentration difference between these regions and the upper troposphere over SYO is relatively large during fall and winter seasons. Therefore, it is probable that this high CO_2 transport is enhanced especially during these seasons. Low CO_2 air is transported to SYO from the mid to high latitude lower troposphere, mainly during fall and winter, as well as from the high latitude low stratosphere all year round; relative absence of transport from the former region in spring and summer reduces the decrease of CO_2 concentration during these seasons, resulting in smaller seasonal amplitude in the upper than in lower atmosphere over SYO. This may be associated with the fact that the vertical concentration difference in high latitude troposphere is small in spring and that the cyclonic activity at the circumpolar zone is suppressed in summer.

As a further investigation of the "origin" of the CO_2 transport to SYO from the southern middle latitudes, a similar back trajectory analysis was carried out for Cape Grim, Tasmania (CGO; 40°41′S, 144°41′E). Results are shown in Fig. 6, which is same as in Fig. 4 but for 350 hPa over CGO. From the comparison with Fig. 5, relatively high CO_2 air reaching 350 hPa over CGO appears to "originate" from regions throughout the tropospheric depth in lower latitudes. Although high CO_2 air which may be associated with high atmospheric convective activity is transported from lower tropospheric regions in lower latitudes during summer and fall, much of the high CO_2 air reaching CGO comes from the upper troposphere. Since the latitudinal gradient of the concentration is steep from late fall to winter, this transport of the high CO_2 air from the upper troposphere is thought to be much enhanced in these seasons. Part of this upper air appears to be transported from the northern hemispheric low latitude region (Fig. 6). Although the ratio of the air parcels transported from this region is small, this transport is likely to influence the concentration variation in the upper troposphere in the SH midlatitudes especially from late fall to winter because the concentration difference between the northern low latitudes and CGO is large in these seasons. This may be the inter-hemispheric transport processes indicated by Nakazawa et al. (1991b). Therefore, it

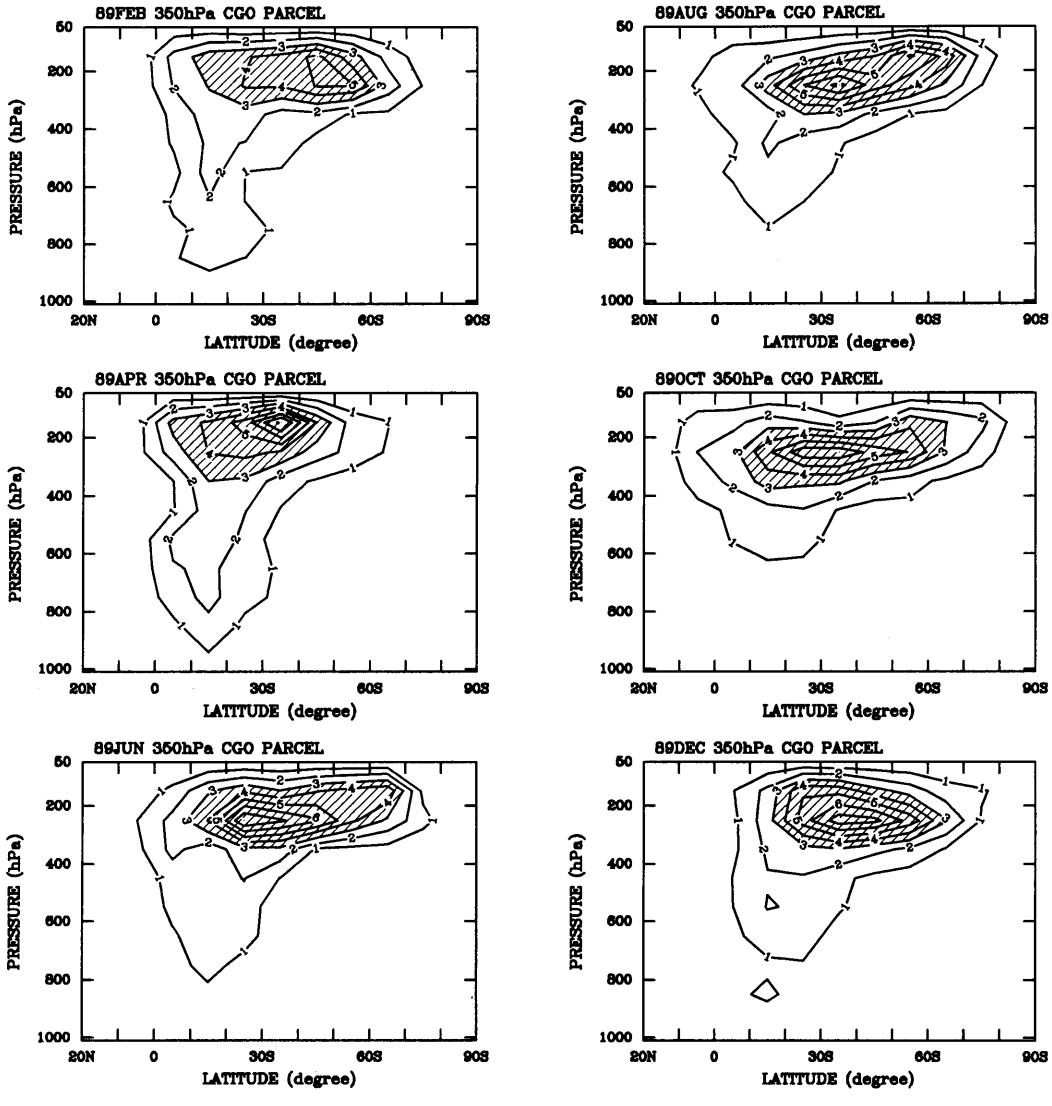

Fig. 6. Same as in Fig. 4 but for 350 hPa over Cape Grim (CGO).

is very possible that part of the air transported to the upper troposphere over SYO originated in the northern hemisphere, supporting the speculation made early in this section. This raises an interesting concept in which the variations of CO_2 concentration observed in the antarctic region are influenced partly by the atmospheric transport of northern hemispheric CO_2 through the upper troposphere.

5. Conclusions

In order to increase our understanding of the seasonal variations in the atmospheric CO_2 concentration over the antarctic region, aircraft and ground-based measurements of the atmospheric CO_2 have been made at SYO since January 1983. The minimum concentration of the average seasonal CO_2 cycle occurs in March throughout

the troposphere over SYO, while the maximum appears in mid-August in the upper troposphere and in late September in the lower and middle troposphere. The peak-to-peak amplitude of the seasonal cycle decreases slightly with height. The CO_2 concentration increases with height during most of the year, but from late winter to spring this height dependency is largely reduced. In order to explore the possibility of contribution of the atmospheric transport processes to these seasonal variations of CO_2 concentration with altitude over SYO, a 3-dimensional trajectory analysis was performed using the US NMC wind data in 1989.

The results of the trajectory analysis indicate the possibility of northern hemispheric air with relatively high CO_2 concentration being transported to SYO through upper troposphere from late fall to winter. This is consistent with the results of the previous atmospheric CO_2 measurements made at SH low and midlatitude regions (Pearman and Beardsmore, 1984; Nakazawa et al., 1991b). A transport of low CO_2 air from SH midlatitudes to SYO along the lower troposphere during spring and summer is also consistent with the seasonal variation of the observed concentration difference between the SH middle and high latitudes (Nakazawa et al., 1991a; Beardsmore and Pearman, 1991a, b).

Along with certain basic assumptions and caveats in the tracing technique in the atmosphere, the trajectory analysis is sensitive to meteorological data. In addition, there are uncertainties associated with the construction of the height-latitude seasonal profiles of the CO_2 concentration field from the data gathered from specific locations in the SH atmosphere. Therefore, the relationship between seasonal transport of seasonally varying CO_2 in the SH troposphere and the average seasonal cycle of CO_2 observed at SYO derived from the trajectory analysis should be interpreted as highly qualitative and suggestive, and certainly not conclusive. For a more definitive and quantitative understanding of the seasonal variation of the CO_2 concentration over Antarctica, further analyses with atmospheric diffusive effect and more accurate 3-dimensional distributions of the atmospheric CO_2 concentration are necessary. Systematic measurements of tropospheric, stratospheric and oceanic CO_2 are required especially in the region from SH middle latitudes to Antarctica where complicated spatial variations of the concentration are present (Pearman and Hyson, 1986; Beardsmore and Pearman, 1987; Conway et al., 1988).

6. Acknowledgements

We express our gratitude to the staffs of the 24, 25, 27, 28, 30, 31 and 32th Japanese Antarctic Research Expedition for their cooperation in collecting air samples. We also thank Dr. Kaz Higuchi for very useful discussions and thoroughly reviewing the manuscript.

7. Appendix

Construction of meridional CO_2 concentration field

The CO_2 concentration fields in height and latitude for individual months of the year were constructed by the following procedures:

The data used were monthly mean values of the CO_2 concentration obtained from the measurements shown in Table 1. All of these data are expressed by the WMO X85 or X81 scale (World Meteorological Organization 1985 or 1981 mole fraction scale) except for our data. Though our calibration scale was developed independent of the WMO scale, it was confirmed from the intercomparison that both scales agree with each other within 0.1 ppmv (Nakazawa et al., 1992). With regard to the Australian data, the possibility of a drift in the calibration scale used was indicated by Pearman et al. (1991) recently. Then, to use the data from the continuous measurements at CGO, the correction was made on the basis of the comparison with the results from the flask sampling measurements by National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostic Laboratory (NOAA/CMDL) at CGO for 1984–1990 (Conway et al., 1990; Beardsmore and Pearman, 1991a).

The best fit curves to these observed data at respective locations were first calculated using a digital-filtering technique (see Subsection 4.1) and then monthly mean values were obtained again from these fit curves. Next, monthly concentration differences between respective locations and SYO were calculated and were averaged for each month of respective years to obtain average monthly

Table 1. *List of the data used for constructing the CO_2 concentration field*

Ground-based continuous measurement

Station	Location	Year	Ref.
South Pole (SPO)	90°S, 25°W	1975–1990	Thoning et al. (1991a); Ferguson (1991)
Syowa Station (SYO)	69°S, 40°E	1984–1992	Nakazawa et al. (1991a, unpublished data)
Cape Grim (CGO)	41°S, 145°E	1976–1990	Beardsmore and Pearman (1991a, b)
Amsterdam Island (AMS)	38°S, 78°E	1980–1990	Gaudry et al. (1991)
American Samoa (SMO)	14°S, 171°W	1976–1990	Thoning et al. (1991b); Ferguson (1991)

Aircraft measurement

Sampling site	Latitude	Height	Year	Flight frequency (per month)	Ref.
Over the Western Pacific Ocean (Tokyo, Japan – Sydney, Australia)	20°N–30°S (every 5°)	10–12 km	1984–1985	1	Nakazawa et al. (1991b)
Over Sydney, Australia	33°S	7.2 km	1984–1985	1	Nakazawa et al. (1991b)
Over Australia	13°S–37°S	8.5 km tropopause	1973–1981	0–6	Pearman and Beardsmore (1984)
Over south-eastern Australia and the Tasman Sea	28°S–43°S	9.0 km tropopause	1972–1979	0–3	Pearman and Beardsmore (1984)
Over Syowa Station	~69°S	0–7 km	1983–1991	0–2	this study

Shipboard measurement

Sampling site	Latitude	Year	Sampling frequency at each latitudinal zone	Ref.
Over the Western Pacific Ocean (Yokohama, Japan – Melbourne, Australia)	20°N–35°S (every 5°) and 39°S	1984–1990	~every 2.5 weeks	Tanaka et al. (1987c); Nakazawa et al., unpublished data

concentration differences from SYO ($\Delta C_{m,a}$), using

$$\Delta C_{m,a} = \frac{1}{n}\sum_i [C_{i,m,a} - C_{i,m,\mathrm{SYO}}], \qquad \text{(A.1)}$$

where $C_{i,m,a}$ is the concentration at the location (a) in the month (m) of the year (i); n is the number of the years used for calculating average monthly difference. In this procedure, we used the data in and after 1984 for SPO, CGO and over the Western Pacific Ocean. For the Australian aircraft data (AUS), since the period covered by measurement is different from that of SYO, the following procedures were performed to obtain average monthly concentration differences from SYO.

(1) The average concentration differences from CGO in each selected month of the year ($\Delta C'_{m,\mathrm{AUS}}$) were calculated from

$$\Delta C'_{m,\mathrm{AUS}} = \frac{1}{n}\sum_i [C_{i,m,\mathrm{AUS}} - C_{i,m,\mathrm{CGO}}]. \qquad \text{(A.2)}$$

(2) From

$$\Delta C''_{m,\mathrm{AUS}} = \Delta C'_{m,\mathrm{AUS}} + \Delta C_{m,\mathrm{CGO}} \approx \Delta C_{m,\mathrm{AUS}}, \qquad \text{(A.3)}$$

the average concentration difference from SYO in the month (m) ($\Delta C_{m,\,\mathrm{AUS}}$) was approximated by $\Delta C''_{m,\,\mathrm{AUS}}$.

(3) These procedures were performed for all months of the year.

For AMS and SMO, similar calculation was made, after calculating average monthly differences from SPO. Then, from

$$\Delta C^{*}_{m,\,a} = \Delta C_{m,\,a} + C_{\mathrm{sea},\,m,\,\mathrm{SYO}}, \qquad (A.4)$$

where $C_{\mathrm{sea},\,m,\,\mathrm{SYO}}$ is the mean concentration in the month (m) of the average seasonal cycle at SYO (Fig. 2), we obtained the deviation from the average concentration at SYO for the period covered by the measurement ($\Delta C^{*}_{m,\,a}$). This was done for all selected locations and months of the year. Since the data over SYO were already represented as the deviations from the average concentration at SYO in Fig. 2, monthly mean values were calculated from the fit curves in the figure.

For the stratospheric data, we have no data from northern low latitudes to the Antarctica. Nakazawa et al. (1991b) suggested, from their lower stratospheric data at northern middle and high latitudes, that the tropospheric CO_2 is injected into the stratosphere mainly by strong convection at low latitudes and then transported poleward. If this transport process occurs symmetrically in both hemispheres, it may be deduced that the annual mean stratospheric CO_2 concentration in the southern hemisphere is comparable to that in the northern hemisphere. In this paper, from the comparison of annual mean concentrations in 1984 and 1985 between lower stratosphere at the northern mid-high latitudes (Nakazawa et al., 1991b) and SYO (Nakazawa et al., 1991a), we assumed that the stratospheric CO_2 concentration at southern mid-high latitudes is lower by 0.48 ppmv than the average concentration at SYO throughout the year. Though the seasonal cycle and height dependency of the CO_2 concentration in the southern stratosphere might be similar to those in the northern hemisphere (Bischof et al., 1985, Gamo et al., 1989, Nakazawa et al., 1991b; 1993b), we neglected them due to lack of observation. However, at least for the transport processes to the lower and middle troposphere over SYO, the influence on the estimated results due to the uncertainty of the stratospheric CO_2 concentration is thought to be small because most of the air parcels arriving at these levels originate in the troposphere. Moreover, since we have no information on height dependency, relative to SYO, of the tropospheric CO_2 over southern higher latitudes, and from the fact that the difference between SYO and SPO is smaller than 0.1 ppmv throughout the year, we assumed that the vertical profiles over Antarctica are the same as over SYO.

Finally, all these data were interpolated linearly in height and latitude to assign monthly CO_2 values to each meridional box (see Section 3). Height-latitude distributions of the CO_2 concentration relative to the average value at SYO in the odd months thus obtained are shown in Fig. 5.

REFERENCES

Beardsmore, D. J. and Pearman, G. I. 1987. Atmospheric carbon dioxide measurements in the Australian region: data from surface observation. *Tellus* **39B**, 42–66.

Beardsmore, D. J. and Pearman, G. I. 1991a. Atmospheric CO_2, Modern record: Cape Grim. In: *Trends '91: A compendium of data on global change* (eds. T. A. Boden, R. J. Sepanski and F. W. Stoss). Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 665 pp.

Beardsmore, D. J. and Pearman, G. I. 1991b. Baseline carbon dioxide concentrations. In: *Baseline atmospheric program* (Australia) 1989 (eds. S. R. Wilson and J. L. Gras). Department of the Arts, Sports, the Environment, Tourism and Territories/ Bureau of Meteorology in cooperation with CSIRO/ Division of Atmospheric Research. Cape Grim BAPS, Tasmania, Australia, 50–51.

Bischof, W., Borchers, R., Fabian, P. and Krüger, B. C. 1985. Increased concentration and vertical distribution of carbon dioxide in the stratosphere. *Nature* **316**, 708–710.

Conway, T. J., Tans, P. P., Waterman, L. S., Thoning, K. W., Masarie, K. A. and Gammon, R. H. 1988. Atmospheric carbon dioxide measurements in the remote global troposphere, 1981–1984. *Tellus* **40B**, 81–115.

Conway, T. J., Tans, P. P. and Waterman, L. S. 1990. Atmospheric CO_2: Cape Grim. In: *Trends '90: A compendium of data on global change* (eds. T. A. Boden, P. Kanciruk and M. P. Farrel). Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 257 pp.

Conway, T. J., Tans, P. P. and Waterman, L. S. 1991. Atmospheric CO_2, modern record. In: *Trends '91: A compendium of data on global change* (eds. T. A.

Boden, R. J. Sepanski and F. W. Stoss). Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 665 pp.

Danielsen, E. F. 1961. Trajectories: Isobaric, isentropic and actual. *J. Meteorol.* **18**, 479–486.

Enting, I. G. and Mansbridge, J. V. 1991. Latitudinal distribution of sources and sinks of CO_2: results of an inversion study. *Tellus* **43B**, 156–170.

Ferguson, E. (ed.) 1991. *Climate monitoring and diagnostic laboratory*. Summary Report 1990, no. 19, ERL, NOAA, Boulder, Colorado, USA, 24–33.

Fung, I., Prentice, K., Matthews, E., Lerner, J. and Russell, G. 1983. Three-dimensional tracer model study of atmospheric CO_2: response to seasonal exchanges with the terrestrial biosphere. *J. Geophys. Res.* **88**, 1281–1294.

Fung, I. Y., Tucker, C. J. and Prentice, K. C. 1987. Application of advanced very high resolution radiometer vegetation index to study atmosphere-biosphere exchange of CO_2. *J. Geophys. Res.* **92**, 2999–3015.

Gamo, T., Tsutsumi, M., Sakai, H., Nakazawa, T., Tanaka, M., Honda, H., Kubo, H. and Itoh, T. 1989. Carbon and oxygen isotopic ratios of carbon dioxide of a stratospheric profile over Japan. *Tellus* **41B**, 127–133.

Gaudry, A., Monfray, P., Polian, G. and Lambert G. 1991. Atmospheric CO_2, modern record: Amsterdam Island. In: *Trends '91: A compendium of data on global change* (eds. T. A. Boden, R. J. Sepanski and F. W. Stoss). Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 665 pp.

Heimann, M., Keeling, C. D. and Tucker, C. J. 1989. A three-dimensional model of atmospheric CO_2 transport based on observed winds: 3. Seasonal cycle and synoptic time scale variations. In: *AGU Monograph* **55**, American Geophysical Union, Washington, USA, 277–303.

Holton, J. R. 1979. *An introduction to dynamic meteorology*. Academic Press, New York, USA, 391 pp.

Inoue, H. and Sugimura, Y. 1988. Distribution and variations of oceanic carbon dioxide in the western North Pacific, eastern Indian, and Southern Ocean south of Australia. *Tellus* **40B**, 308–320.

Keeling, C. D., Adams, J. A. Jr., Ekdahl, C. A. Jr. and Guenther, P. R. 1976. Atmospheric carbon dioxide variations at the South Pole. *Tellus* **28**, 552–564.

Keeling, C. D., Bacastow, R. B., Carter, A. F., Piper, S. C., Whorf, T. P., Heimann, M., Mook, W. G. and Roeloffzen, H. 1989a. A three-dimensional model of atmospheric CO_2 transport based on observed winds: 1. Analysis of observational data. In: *AGU Monograph* **55**, American Geophysical Union, Washington, USA, 165–236.

Keeling, C. D., Piper, S. C. and Heimann, M. 1989b. A three-dimensional model of atmospheric CO_2 transport based on observed winds: 4. Mean annual gradients and interannual variations. In: *AGU Monograph* **55**, American Geophysical Union, Washington, USA, 305–362.

Keeling, C. D. 1991. Atmospheric CO_2, modern record. In: *Trends '91: A compendium of data on global change* (eds. T. A. Boden, R. J. Sepanski and F. W. Stoss). Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 665 pp.

Komhyr, W. D., Gammon, R. H., Harris, T. B., Waterman, L. S., Conway, T. J., Taylor, W. R. and Thoning, K. W. 1985. Global atmospheric CO_2 distribution and variations from 1968–1982 NOAA/GMCC CO_2 flask sample data. *J. Geophys. Res.* **90**, 5567–5596.

Merrill, J. T. 1989. Atmospheric long-range transport to the Pacific Ocean. In: *Chemical oceanography* (eds. J. P. Riley, R. Chester and R. A. Duce). Academic Press, London, UK, 15–50.

Nakazawa, T., Aoki, S., Murayama, S., Fukabori, M., Yamanouchi, T., Murayama, H., Shiobara, M., Hashida, G., Kawaguchi, S. and Tanaka, M. 1991a. The concentration of atmospheric carbon dioxide at Japanese Antarctic Station, Syowa. *Tellus* **43B**, 126–135.

Nakazawa, T., Miyashita, K., Aoki, S. and Tanaka, M. 1991b. Temporal and spatial variations of upper tropospheric and lower stratospheric carbon dioxide. *Tellus* **43B**, 106–117.

Nakazawa, T., Murayama, S., Miyashita, K., Aoki, S. and Tanaka, M. 1992. Longitudinally different variations of lower tropospheric carbon dioxide concentrations over the North Pacific Ocean. *Tellus* **44B**, 161–172.

Nakazawa, T., Morimoto, S., Aoki, S. and Tanaka, M. 1993a. Time and space variations of the carbon isotopic ratio of tropospheric carbon dioxide over Japan. *Tellus* **45B**, 258–274.

Nakazawa, T., Machida, T., Murayama, S., Hashida, G., Morimoto, S., Tanaka, M., Honda, H. and Itoh, T. 1993b. Concentration variations of stratospheric carbon dioxide over Japan. In: *Proceedings WMO/GAW 4th International CO_2 Conference*. Carqueiranne, France, 13–17 September 1993. Environmental Pollution Monitoring and Research Programme Report No. 89, 20–23.

Pearman, G. I. and Beardsmore, D. J. 1984. Atmospheric carbon dioxide measurements in the Australian region: ten years of aircraft data. *Tellus* **36B**, 1–24.

Pearman, G. I. and Hyson, P. 1986. Global transport and inter-reservoir exchange of carbon dioxide with particular reference to stable isotope distributions. *J. Atmos. Chem.* **4**, 81–124.

Pearman, G. I., Beardsmore, D. J. and Francey, R. J. 1991. Zonal representativeness of Cape Grim CO_2 measurements. In: *Climate monitoring and diagnostic laboratory*, Summary Report 1990, no. 19, (ed. E. Ferguson). ERL, NOAA, Boulder, Colorado, USA, 107–108.

Taguchi, S. 1994. Cross-tropical trajectories in the troposphere. *J. Meteorol. Soc. Japan* **72**, 531–553.

Tanaka, M., Nakazawa, T., Shiobara, M., Ohshima, H., Aoki, S., Kawaguchi, S., Yamanouchi, T., Makino, Y. and Murayama, H. 1987a. Variation of atmospheric carbon dioxide concentration at Syowa Station (69°00′S, 39°35′E), Antarctica. *Tellus* **39B**, 72–79.

Tanaka, M., Nakazawa, T. and Aoki, S. 1987b. Time and space variations of tropospheric carbon dioxide over Japan. *Tellus* **39B**, 3–12.

Tanaka, M., Nakazawa, T. and Aoki, S. 1987c. Seasonal and meridional variations of atmospheric carbon dioxide in the lower troposphere of the northern and southern hemispheres. *Tellus* **39B**, 29–41.

Tans, P. P., Conway, T. J. and Nakazawa, T. 1989. Latitudinal distribution of the sources and sinks of atmospheric carbon dioxide derived from surface observations and an atmospheric transport model. *J. Geophys. Res.* **94**, 5151–5172.

Tans, P. P., Fung, I. Y. and Takahashi, T. 1990. Observational constraints on the global atmospheric CO_2 budget. *Science* **247**, 1431–1438.

Thoning, K. W., Gillette, D. A., Komhyr, W. D., Tans, P. P. and Waterman, L. S. 1991a. Atmospheric CO_2, modern record: Amundsen Scott. In: *Trends '91: A compendium of data on global change* (eds. T. A. Boden, R. J. Sepanski and F. W. Stoss). Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 665 pp.

Thoning, K. W., Tans, P. P., Waterman, L. S., Komhyr, W. D., Nelson, D. W. and Harris, T. B. 1991b. Atmospheric CO_2, modern record: Cape Matatula. In: *Trends '91: A compendium of data on global change* (eds. T. A. Boden, R. J. Sepanski and F. W. Stoss). Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 665 pp.

Trenberth, K. E. and Olson, J. G. 1988. An evaluation and intercomparison of global analyses from the National Meteorological Center and the European Center for Medium Range Weather Forecasts. *Bull. Am. Meteorol. Soc.* **69**, 1047–1057.

Yamazaki, K., Okada, K. and Iwasaka, Y. 1989. Where do aerosol particles in the antarctic upper troposphere come from? (a case study in January 1983). *J. Meteorol. Soc. Japan* **67**, 889–906.

Yamazaki, K. 1992. The statistical analysis of long-term trajectory calculation based on the observed data. *J. Meteorol. Soc. Japan* **70**, 1167–1173.