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Time and space variations of tropospheric carbon dioxide over Japan

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ABSTRACT

Aircraft measurements of atmospheric CO_2 concentration over Japan, initiated in January 1979, have been continued to the present. The average seasonal variation of atmospheric CO_2 showed maximum concentration early in April and early in May, and minimum concentration in mid-August and mid-September for the lower-most and the upper-most layers of the troposphere, respectively. The peak-to-peak amplitudes of the seasonal variation were 14.5, 9.0 and 7.8 ppmv for the lower, middle and upper tropospheres, respectively. The average rate of annual increase of the CO_2 concentration over the last 6 years was about 1.3 ppmv yr^{-1} with considerable variation with time. The vertical profile of the annual mean value of the CO_2 concentration was almost the same from year to year; the CO_2 concentration decreased gradually with height and the concentration difference between the lowest and highest layers of the troposphere was about 2 ppmv.

1. Introduction

The number of measurements of atmospheric CO_2 concentration have been increasing rapidly in recent years; approximately 40 ground stations are currently operating around the world (Pearman, 1980). Because the sources and sinks of atmospheric CO_2 are on the earth's surface, the temporal variation of the CO_2 concentration is most pronounced in the lowest layer of the atmosphere and is reduced gradually with height due to enhanced mixing of airmasses of different origins (Bolin and Bischof, 1970; Tanaka et al., 1983a). The horizontal variability of the CO_2 concentration over a distance of a few hundred km or more is also reduced greatly in the middle and upper tropospheres with respect to the surface (Bolin and Bischof, 1970; Pearman and Garratt, 1973; Tanaka et al., 1983a). An aircraft measurement is therefore one of the most promising methods of detecting time and space variations of the concentration of atmospheric CO_2 (Keeling et al., 1968; Bolin and Bischof, 1970; Tanaka et al., 1983a; Pearman and Beardsmore, 1984).

Our aircraft measurements were initiated in January 1979 to elucidate temporal and spatial variations of the tropospheric CO_2, and to contribute to better understanding of the global carbon cycle. In this paper, we report the results of our measurements for the period January 1979–May 1985.

2. Experimental procedures

Details of our experimental procedures have been described elsewhere (Tanaka et al., 1983a, b, 1987). Only a brief descriptional and supplemental explanations will be presented here.

2.1. Air sampling and analysis

The aircraft used for collecting air samples were a Piper Cherokee 140-C (Japan Flying Service) and a McDonell Douglas DC-9-41 (Toa Domestic Airlines). Each air sample was collected, one at a time, into a 550 ml Pyrex glass

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flask by pressurizing up to about 3 atm with an electric rubber-diaphragm or manual stainless-steel-plunger pump. The air sample was taken from the intake mounted in a gap between the propeller and the engine cowling for the Piper and from the so called "fresh air" nozzle for the DC-9. Air sampling on the DC-9 was made in the cabin in the early part of the program, but in the cockpit from 1981 to more precisely know the location and altitude at which the air sample was taken.

Air samples collected in the flasks were returned to our laboratory and analyzed by a nondispersive infrared analyzer within a day after sampling to avoid the deterioration of air samples. Water vapor contained in the air samples was removed by 2 glass traps cooled at -78°C. The calibration of the analyzer response was made by introducing 3 or 4 standard gases into the sample cell about once per hour. The overall imprecision of our flask analyses was ± 0.1 ppmv.

2.2. *Standard gases*

Our standard gases were CO_2-in-air mixtures, and were classified into 3 categories, primary, secondary and working (Tanaka et al., 1987). The working and secondary standard gases were stored in 47 l manganese-steel cylinders, and the primary standard gases in 10 l manganese-steel or aluminum cylinders. The CO_2 concentration of the working standard gases was calibrated before and after their use against the secondary standard gas system to confirm the drift in concentration to be almost within ± 0.1 ppmv as shown in Tanaka et al. (1987). The secondary standard gases were compared with the primary standard gases twice or three times per annum. The lifetimes of the working standards were about 2 years, while those of the secondary standards ranged between 4 and 6 years.

The primary standard gases were prepared gravimetrically by using a 2-stage dilution and 10 l manganese-steel cylinders at the beginning of our program (Tanaka et al., 1983b). The uncertainties in their CO_2 concentration were estimated to be ± 0.3–0.4 ppmv from uncertainty of ± 25 mg yielded in weighing each component gas on the balance. To minimize uncertainties in calibrated values, CO_2 concentrations of the primary standards were corrected by a least-square-fit technique assuming a quadratic relation between the response of our analyzer and the CO_2 concentration, as shown in Table 1. Our primary standard gases were prepared again in 1981 by the same procedures. The result of comparison showed that the standard-gas scales established in 1979 and 1981 were consistent with each other within estimated uncertainties. However, considerable efforts were also continued from that time to reduce uncertainties in the standard-gas scale. In 1983 and 1985, primary

Table 1. *Intercomparison of the primary standard gases prepared in 1979, 1981, 1983 and 1985; values in parentheses were obtained by a least-square-fit correction assuming a quadratic relation between the response of the analyzer and the CO_2 concentration*

1979		
Cylinder	CO_2 (ppmv)	
4K-12313	310.3 (310.27)	
1K-44382	331.1 (331.25)	
1K-44342	346.3 (346.12)	
3K-31033	359.3 (359.37)	
1981		
Cylinder	CO_2 (ppmv)	Calibration by the 1979 standards
3K-49048	310.8 (310.71)	310.78
3K-41954	318.4 (318.54)	318.70
CX-84702	330.0 (330.02)	330.19
CX-55802	338.9 (338.86)	338.97
CX-52083	349.4 (349.31)	349.24
3K-98609	359.5 (359.56)	359.23
1983		
Cylinder	CO_2 (ppmv)	Calibration by the 1979 standards
DC4753	320.76 (320.77)	321.28
DC4754	329.81 (329.81)	330.40
DC4755	339.74 (339.73)	340.25
DC4756	350.12 (350.11)	350.58
DC4757	359.49 (359.53)	359.87
DC4758	368.26 (368.24)	
1985		
Cylinder	CO_2 (ppmv)	Calibration by the 1983 standards
DC7338	320.02 (320.04)	320.04
DC7339	328.92 (328.89)	328.87
DC7340	341.00 (340.97)	340.91
DC7341	350.45 (350.49)	350.39
DC7342	360.95 (360.97)	360.82
DC7343	369.64 (369.62)	369.41

standard gases were prepared repeatedly by adopting a 3-stage dilution and 10 l aluminum cylinders (Tanaka et al., 1987). The imprecision (one standard deviation) in the CO_2 concentration of these gases were estimated to be ± 0.13 ppmv. Both scales of 1983 and 1985 agreed within estimated uncertainties of respective scales, but were systematically lower by about 0.5 ppmv than the previous scales (Table 1). The reason for this large discrepancy exceeding the limit of estimated uncertainties of the respective scales is unsolved yet. However, since great care was taken in preparing the primary standard gases in 1983 and 1985, we adopted the 1983 scale for our standard gases in June 1985.

3. Results and discussion

The data obtained were first grouped into 5 height intervals of 0–2, 2–4, 4–6, 6–8 and 8 km-tropopause, and then, measured values of the CO_2 concentration included in the respective intervals were simply averaged. The stratospheric data as well as some anomalous data observed occasionally especially in the lower troposphere (Tanaka et al., 1983a) were excluded from this data processing. The averaged values of the CO_2 concentration, thus obtained, are shown in Fig. 1 and Table 2. To characterize the temporal variations of the atmospheric CO_2 concentration over Japan, a spectral analysis was applied to these data set by adopting the maximum entropy method (Burg, 1967). Since the algorithm required equally spaced data, the CO_2 concentration of the 15th of each month was interpolated from contiguous data subjectively by drawing the best fit curve by hand. A power spectrum obtained for the upper-most troposphere is given in Fig. 2 as an example of the results. The harmonics significantly contributing to the long-term variations of tropospheric CO_2 over Japan were the fundamental annual cycle and its first harmonics. The results for the lower height intervals were very similar to this, except that weak 2nd and 3rd harmonics which appear in Fig. 2, are a little more prominent in the lowermost layers.

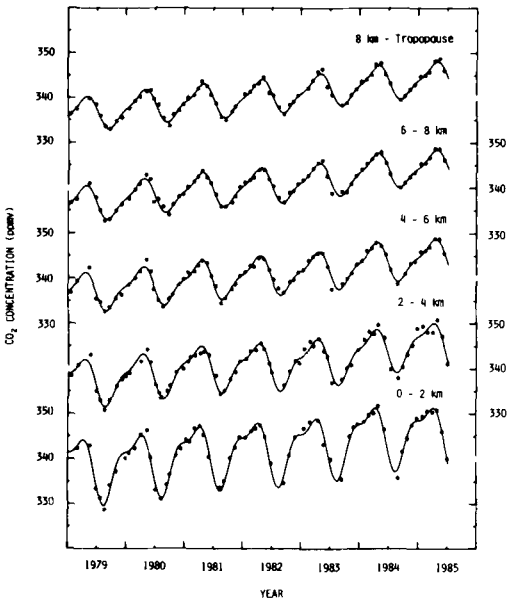

Fig. 1. Concentrations of atmospheric CO_2 for respective height intervals of the troposphere over Japan. Circles represent average values of the measured CO_2 concentrations and curves the best fit function to the data.

We therefore separated the seasonal and secular variations of the CO_2 concentration by fitting the observed values to Fourier harmonics,

$$S(t) = C_1 \sin 2\pi t + C_2 \cos 2\pi t + C_3 \sin 4\pi t + C_4 \cos 4\pi t, \quad (1)$$

and cubic splines with yearly knots, representing respective contributions, by using an iterative procedure. Here, t is the elapsed time (years) relative to January 1, 1979 and the C_i's are parameters determined by the fitting. The standard deviations of the observed values from the fitted functions were 0.54, 0.62, 0.69, 0.99 and 1.03 ppmv for the height intervals of tropopause—8, 8–6, 6–4, 4–2 and 2–0 km, respectively. The average seasonal CO_2 variations are shown in Fig. 3. In general, the seasonal variation decreases and its phase is delayed markedly with increasing height; the maximum concentration appeared early in April and early in May, and the maximum concentration in mid-August and mid-September in the lowest and the highest layers of the troposphere, respectively, and peak-to-peak amplitudes for the lower, middle and upper tropospheres were 14.5, 9.0 and 7.8 ppmv. Fig. 3 also shows the CO_2 concentration increase with

Table 2. *Concentrations of atmospheric CO_2 (in ppmv above 300.0) in respective height intervals of the troposphere and in the lower stratosphere over Japan; numbers in parentheses indicate the date of air sampling, number of samples and the standard deviation, respectively*

	0 – 2 km						
	1979	1980	1981	1982	1983	1984	1985
Jan		41.42 (21; 6;0.31)	44.28 (14;20;1.04)	44.86 (21;11;0.10)	47.02 (20;10;0.16)	48.39 (30;11;0.32)	49.62 (30;10;0.11)
Feb		42.40 (26; 1; –)	44.02 (5;20;0.34)	46.37 (26;10;0.19)	48.43 (26;10;0.11)	49.82 (28;10;0.12)	50.77 (27;11;0.48)
Mar	42.43 (3; 5;0.54)		46.92 (7;10;1.07)	46.92 (24; 9;0.23)		50.23 (31; 9;0.47)	
Apr		45.53 (3;10;0.46)	47.38 (9;10;2.15)	47.60 (23;11;0.96)	48.78 (21;11;0.78)	51.83 (26; 7;0.95)	50.48 (3; 8;0.68)
May	43.06 (21; 5;0.66)	46.42 (12; 8;1.23)	45.38 (2;11;0.71)	45.11 (22;11;0.32)	43.36 (22; 9;0.40)		50.95 (1; 8;0.67)
Jun	33.45 (26; 5;1.10)	40.38 (4; 4;0.86)	40.56 (3;12;1.23)	39.20 (30;12;0.68)		46.68 (6;10;0.95)	46.20 (1; 7;1.27)
Jul	31.37 (24; 2;0.95)	33.22 (4;11;2.89)			40.11 (7;11;2.21)	37.34 (1;10;1.62)	40.19 (3; 5;1.06)
Aug	28.77 (20; 6;1.52)	31.95 (8;13;3.02)	33.80 (13;10;1.30)			35.96 (29;10;0.55)	
Sep	34.32 (21; 4;1.31)	34.46 (12;21;0.55)	35.16 (8; 8;0.50)	34.78 (17;11;0.92)	35.62 (14;11;0.83)	41.80 (26;10;0.18)	
Oct		36.72 (1;21;0.44)	40.42 (15;11;1.47)	40.93 (21;11;0.42)	45.26 (31;11;1.50)	44.71 (26;10;1.03)	
Nov	37.32 (1; 2;0.40)	40.94 (10;21;0.82)	42.64 (19;11;2.38)	44.93 (26;10;0.10)	47.26 (25;10;0.25)	47.70 (26;11;0.73)	
Dec	40.26 (28; 6;0.31)	42.37 (11; 9;0.23)	44.98 (12;11;0.29)		47.84 (22;11;3.54)	49.19 (26;11;0.30)	

	2 – 4 km						
	1979	1980	1981	1982	1983	1984	1985
Jan	38.49 (19; 1; –)	39.10 (23; 9;1.12)	41.70 (23; 1; –)	42.23 (21;15;0.88)	44.58 (20; 1; –)	46.54 (30; 9;0.15)	49.55 (30;10;0.15)
Feb			42.68 (5; 8;0.95)	44.01 (16; 2;0.42)	46.14 (26;10;1.88)	48.33 (28;10;1.21)	48.22 (27; 9;1.11)
Mar	40.85 (3; 4;0.93)		43.06 (7; 5;0.57)	44.22 (24; 9;1.02)	45.13 (19;11;0.64)	47.79 (31; 8;0.76)	
Apr		41.65 (3; 5;1.07)	43.46 (9; 8;0.81)	45.69 (23; 7;0.73)	46.75 (21; 9;1.51)	49.84 (26; 8;0.88)	48.15 (3; 8;0.07)
May	43.17 (21; 5;0.57)	44.28 (12; 7;0.58)	43.83 (2; 7;0.48)	44.50 (13; 4;0.17)	44.01 (22; 8;0.67)		50.98 (1; 8;1.00)
Jun	35.08 (26; 6;0.36)	41.50 (4; 6;1.40)	43.09 (3; 8;0.27)	41.20 (15; 4;0.28)	42.83 (14; 2;0.48)	46.99 (6;10;0.46)	47.24 (1; 8;0.51)
Jul	33.08 (24; 3;1.38)	36.50 (4; 8;1.20)	38.51 (17; 1; –)	39.26 (1; 9;0.66)	37.05 (13; 1; –)	37.83 (1; 7;1.20)	41.20 (3; 8;1.01)
Aug	30.89 (20; 6;0.85)	33.57 (8; 7;0.43)	34.46 (13; 7;1.25)			38.04 (29; 8;0.83)	
Sep	33.05 (21; 6;0.32)	35.03 (12; 8;0.30)	35.25 (8; 8;0.50)	36.46 (17;12;0.67)	37.87 (14; 7;0.49)	40.52 (26; 8;0.47)	
Oct	36.21 (31; 6;0.73)	36.30 (1; 8;0.27)	37.82 (15; 8;0.70)	39.55 (21; 8;0.73)	40.31 (14; 3;0.20)	43.30 (26; 8;0.36)	
Nov	37.75 (30; 1; –)	39.29 (10; 9;1.14)	39.36 (19;11;0.13)	41.84 (26;15;1.96)	40.93 (11; 2;0.01)	45.36 (26;11;0.69)	
Dec	38.33 (28; 5;1.44)	39.99 (25; 2;0.12)	41.74 (12; 4;1.46)	41.32 (20; 8;0.22)	44.28 (22; 8;0.52)	49.07 (26; 8;0.91)	

4 – 6 km

	1979	1980	1981	1982	1983	1984	1985
Jan	36.96 (19; 3;0.49)	38.70 (21; 7;0.85)	41.42 (23;10;0.24)	41.52 (14; 6;0.83)	42.11 (13;10;0.41)	44.07 (12; 7;0.31)	46.09 (16; 7;0.32)
Feb	39.51 (27; 3;1.07)	40.12 (26; 4;0.25)	41.70 (28; 5;0.40)	42.92 (16; 8;0.54)	44.16 (15;10;0.81)	46.32 (14; 5;0.70)	46.33 (15; 2;0.09)
Mar		41.62 (28; 4;0.36)	42.99 (28; 1; –)	42.68 (13; 7;0.49)	44.99 (15; 4;0.74)	46.86 (16; 7;0.70)	47.09 (12; 6;0.32)
Apr		44.21 (30; 1; –)	44.07 (21; 1; –)	44.56 (13; 9;0.43)	45.72 (15; 8;0.76)	48.04 (12; 5;0.41)	49.01 (15; 6;1.23)
May	42.52 (15; 4;0.61)	41.64 (31; 1; –)	43.49 (22; 7;0.61)	44.39 (13; 8;0.33)	45.74 (14;10;0.22)	47.35 (15; 6;0.20)	48.89 (14; 7;0.51)
Jun	35.57 (26; 5;0.74)	37.55 (25; 4;1.06)	40.66 (17; 9;1.28)	42.01 (15; 4;0.20)	42.69 (14; 9;0.41)	45.43 (13; 5;0.64)	45.72 (14; 6;0.61)
Jul	34.79 (24; 1; –)	35.19 (23; 5;0.42)	38.34 (17; 5;0.31)	39.87 (13; 6;0.98)	37.71 (13;11;1.27)	42.12 (13; 5;1.54)	
Aug	32.73 (20; 5;0.64)	33.81 (22; 6;2.15)	34.66 (20; 5;0.70)	37.92 (15; 1; –)		38.97 (25; 2;0.18)	
Sep	33.71 (21; 4;0.33)	35.61 (26; 6;0.36)	35.85 (18; 9;0.29)	36.96 (17; 9;0.44)	39.02 (13; 1; –)	39.01 (14; 1; –)	
Oct		36.66 (22; 5;0.16)	37.70 (27; 5;0.17)	39.28 (20; 8;0.62)	40.16 (14;10;0.58)	41.15 (13; 7;0.42)	
Nov	35.69 (1; 2;0.93)	38.73 (29; 4;0.25)	38.82 (17; 6;0.56)	39.76 (15;10;0.50)	41.33 (11;11;0.70)	43.46 (13; 7;0.67)	
Dec	36.39 (4; 5;0.84)	39.90 (25; 1; –)	40.69 (14;10;0.24)	41.12 (13;12;0.30)	43.28 (12;10;0.55)	43.91 (12; 6;0.18)	

6 – 8 km

	1979	1980	1981	1982	1983	1984	1985
Jan	36.75 (19; 1; –)	37.69 (21; 8;0.47)	40.27 (23; 2;0.47)	41.31 (14;10;0.32)	41.88 (13;12;0.45)	44.23 (12; 6;0.33)	45.55 (16; 7;0.34)
Feb	37.65 (27; 9;0.49)	39.66 (26; 5;0.59)	41.45 (28; 8;0.29)	41.87 (16;11;0.45)	42.87 (15;13;0.47)	45.55 (14; 8;0.49)	45.66 (15; 4;0.39)
Mar		40.87 (28; 6;0.44)	42.53 (28;12;0.62)	42.41 (13; 9;0.23)	44.45 (15;13;0.61)	46.55 (16; 7;0.75)	46.88 (12; 9;0.42)
Apr		42.97 (30; 6;1.33)	43.90 (21;10;1.13)	44.06 (13;10;0.34)	45.64 (15;12;0.55)	37.53 (12; 9;0.31)	48.88 (15; 7;0.75)
May	41.10 (15; 1; –)	41.99 (31; 6;0.17)	42.38 (22;11;0.42)	44.08 (13;15;0.21)	46.23 (14;12;0.57)	48.22 (15; 7;0.73)	48.79 (14; 6;0.41)
Jun	37.93 (26; 3;0.45)	36.92 (25; 4;0.45)	41.08 (17;11;0.28)	41.97 (15; 4;0.44)	42.68 (14;13;0.45)	45.69 (13; 7;0.21)	46.32 (14; 7;0.65)
Jul	35.08 (24; 5;0.85)	37.53 (23; 1; –)	38.61 (17;15;0.34)	40.46 (13; 7;0.60)	38.89 (13; 7;0.91)	43.16 (13; 5;0.68)	
Aug	32.85 (20; 8;0.43)	35.83 (22; 1; –)	35.97 (20; 7;0.32)	37.92 (15; 7;0.27)			
Sep	33.15 (21; 7;0.24)	34.06 (26; 1; –)	35.93 (18; 6;0.44)	36.83 (17;13;0.63)	39.37 (13; 8;0.28)	40.21 (14; 7;0.62)	
Oct		36.40 (22; 2;0.06)	36.83 (27;15;0.74)	38.99 (20;11;0.65)	39.13 (14;13;0.43)	41.20 (13; 6;0.37)	
Nov	35.04 (1; 2;0.21)	38.17 (29;14;0.46)	38.19 (17;14;0.31)	39.41 (15;11;0.48)	40.93 (11;12;0.41)	42.54 (13; 9;0.87)	
Dec	36.39 (4; 8;0.99)	38.58 (25; 5;0.54)	40.29 (14; 9;0.81)	40.88 (13;16;0.47)	42.74 (12;12;0.32)	43.56 (12; 7;0.04)	

8 km – Tropopause

	1979	1980	1981	1982	1983	1984	1985
Jan	36.54 (19; 3;0.27)	37.69 (21; 1; –)	40.15 (23; 1; –)	40.93 (14;14;0.37)	41.53 (13; 1; –)	43.78 (12; 7;0.51)	44.97 (16; 5;0.19)
Feb	37.65 (27; 1; –)	39.38 (26; 7;0.27)	40.79 (28;20;0.28)	41.48 (16;13;0.31)	42.36 (15;12;0.25)	44.31 (14; 5;0.53)	45.15 (15; 5;0.37)
Mar		40.29 (28; 9;0.35)	42.23 (28; 7;0.37)	42.73 (13;12;1.11)	44.03 (15;11;0.52)	45.32 (16;10;0.76)	45.85 (12; 6;0.29)
Apr		41.55 (30; 9;0.92)	43.78 (21; 8;1.45)	43.33 (13;17;0.91)	45.93 (15; 8;0.43)	47.58 (12; 6;0.37)	48.38 (15; 8;0.59)
May	39.97 (15;11;0.68)	41.83 (31;14;0.56)	42.69 (22;14;0.45)	44.74 (13;13;0.43)	46.51 (14;13;0.49)	47.83 (15; 9;0.46)	48.85 (14; 9;0.31)
Jun	48.59 (26;14;0.42)	39.75 (25; 8;0.57)	40.76 (17;11;0.48)	41.22 (15;20;0.87)	42.62 (14; 9;0.47)	45.36 (13;10;0.57)	46.21 (14;10;0.49)
Jul	36.02 (24;17;0.59)	38.50 (23;14;0.63)	38.86 (17;19;0.76)	40.64 (13;19;0.59)	40.71 (13; 6;0.49)	43.37 (13; 7;0.61)	
Aug	33.77 (20; 7;1.10)	35.49 (22;12;0.62)	35.92 (20;23;1.15)	37.89 (15; 5;0.12)			
Sep	33.09 (21;12;0.37)	33.86 (26;18;1.09)	35.18 (18;24;0.40)	36.46 (17;10;0.63)	38.43 (13; 5;0.30)	39.65 (14;10;0.40)	
Oct		36.36 (22; 7;0.15)	37.13 (27;19;0.30)	38.48 (20;14;0.50)	38.96 (14;10;0.37)	40.54 (13; 7;0.52)	
Nov	34.84 (1;12;0.44)	37.30 (29; 9;0.18)	38.14 (17;15;0.13)	39.19 (15;12;0.50)	40.83 (11;11;0.26)	42.06 (13; 7;0.52)	
Dec	35.58 (4; 9;0.47)	38.58 (25; 8;0.17)	39.45 (14; 1; –)	40.64 (13; 5;0.42)	42.04 (12;10;0.42)	43.09 (12;10;0.31)	

Lower stratosphere

	1979	1980	1981	1982	1983	1984	1985
Jan	34.72 (19; 8;0.49)	36.45 (21; 5;0.24)	38.33 (23; 1; –)	38.75 (14; 5;0.03)	40.06 (13; 8;0.28)	42.21 (12; 3;0.20)	43.54 (16; 3;0.23)
Feb	34.01 (27; 4;0.17)			39.72 (16; 6;0.13)		41.98 (14; 4;0.46)	43.45 (15; 6;0.61)
Mar			37.77 (28; 4;0.15)	39.84 (13; 6;0.24)			
Apr		36.33 (30; 2;0.39)	38.18 (21; 1; –)				44.14 (15; 2;0.75)
May							
Jun							
Jul							
Aug							
Sep							
Oct							
Nov							
Dec		38.04 (25; 8;0.32)	38.61 (14;18;0.31)				

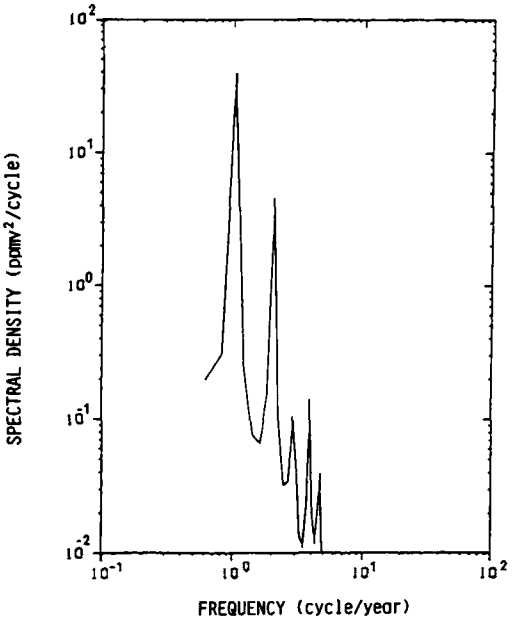

Fig. 2. Power spectrum of CO_2 concentrations observed in the height interval of 8 km-tropopause over Japan for the period January 1979–December 1984.

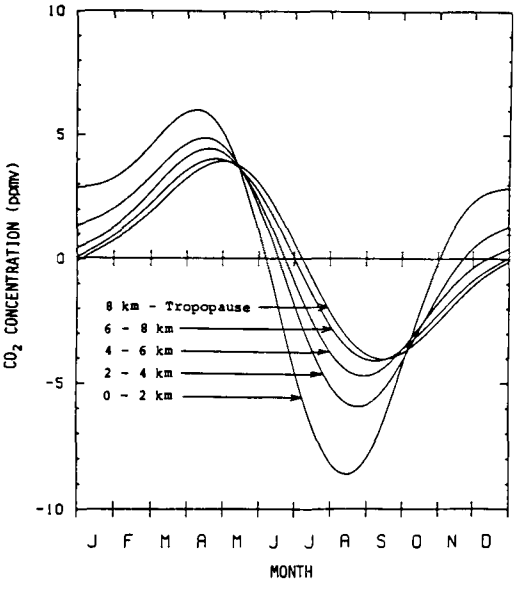

Fig. 3. Average seasonal variations of atmospheric CO_2 concentration over Japan.

height for the period from June to September and vice versa for the remaining period, reflecting the effect of biospheric activities in the northern hemisphere. Such a seasonal variation of tropospheric CO_2 is closely similar to that observed in high latitudes of the northern hemisphere over the Atlantic Ocean (Bolin and Bischof, 1970).

Fig. 4 shows the secular trend and the rate of annual increase of the CO_2 concentration for the upper-most layer of the troposphere. The rate of annual increase was found to be about 1.3 ppmv yr^{-1} as an average of the last 6 years, which is in excellent agreement with the result of mid-tropospheric data collected over south-eastern Australia for 1972–1981 (Pearman and Beardsmore, 1984). However, detailed inspection of the results showed that the rate was decreased to 1.0 ppmv yr^{-1} from the middle of 1981 to the middle of 1982, increased to 1.8 ppmv yr^{-1} in late 1983, and decreased again to less than 1.0 ppmv yr^{-1} in 1984. Similar trends were also observed for other height intervals. The values of the increasing rate in the beginning of 1979 and in 1985 are not credible due to the unconstrained nature of the first and last segments of the cubic spline for knots at both ends of the record. The rate of change of atmospheric CO_2 concentration has been shown to correlate with the Southern Oscillation Index (SOI), and consequently with El Niño occurrences (Bacastow, 1976; Bacastow et al., 1980; Bacastow and Keeling, 1981). The time lags to attain maximum correlations were found to be 6, 1–2, 3 and 6–7 months for the South Pole, Fanning Island, Mauna Loa and Station P, respectively, with respect to the SOI (Bacastow et al., 1980; Bacastow and Keeling, 1981). The enhanced rate of increase of the CO_2 concentration over Japan in 1983 may also be related to the 1982/1983 ENSO event. The time lag was estimated to be 6–8 months for the uppermost layer of the troposphere over Japan by comparing with 5-month running means of the SOI which shows the maximum value in mid-1981 and the minimum value in January of 1983 (Bergman, 1984). The lags for other height intervals also ranged between 6 and 10 months, and no definite variation with height was observed except that the minimum growth rate was apt to appear in early 1982 in the lower layers. The time lag of around 8 months is larger by a few months than expected from the results of the previous

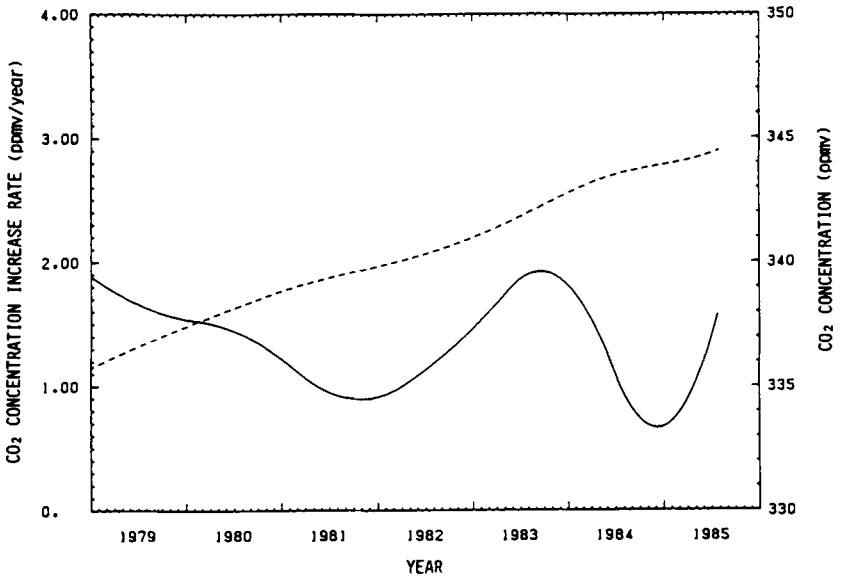

Fig. 4. Secular trend (dashed curve) and rate of annual increase (solid curve) of the atmospheric CO_2 concentration observed in the upper-most layer of the troposphere over Japan.

Table 3. *Fossil fuel airborne fractions for 1979–1982*

Year	CO_2 concentration increase (ppmv yr^{-1})							Airborne Fraction
	0 – 2 km	2 – 4 km	4 – 6 km	6 – 8 km	8 km – Trop.	Average	Emission	
1979 – 1980	1.38	1.42	1.33	1.53	1.55	1.44	2.51	0.57
1980 – 1981	1.75	1.52	1.41	1.37	1.23	1.46	2.46	0.59
1981 – 1982	0.77	0.53	0.91	0.97	0.98	0.83	2.42	0.34
Mean	1.30	1.16	1.22	1.29	1.25	1.24	2.46	0.50

studies. In this respect, the latest data of the NOAA/GMCC Flask Network showed that the maximum value of the globally averaged CO_2 growth rate appeared in the third to fourth quarter of 1983, similar to our results (NOAA/GMCC, 1986). The above data also showed, however, that the minimum growth rate appeared in mid-1982, later by a few or several months than ours. More careful and extended analysis than ours are thought to be necessary to establish the correlation between the SOI and anomalies in the CO_2 growth rate.

It is well-known that the airborne fractions of anthropogenically injected CO_2 into the atmosphere are highly variable from year-to-year (Keeling and Bacastow, 1977; Pearman and Beardsmore, 1984; Komhyr et al., 1985). Komhyr et al. (1985) reported that the airborne fractions calculated from globally-averaged values of the atmospheric CO_2 concentration are 0.81, 0.49 and 0.30 on January 1 of 1980, 1981 and 1982, respectively. Table 3 shows the airborne fractions estimated from our data and the data of fossil fuel combustion and cement manufacture (Marland and Rotty, 1984), assuming a global atmospheric mass of 5.12×10^{18} kg (Trenberth, 1981) and a mean molecular weight of 28.97 (NOAA/NASA/USAF, 1976). Our values are different from those of Komhyr et al. (1985) especially for 1980 and 1981, but the averaged airborne fractions over the same period are in good agreement with each other.

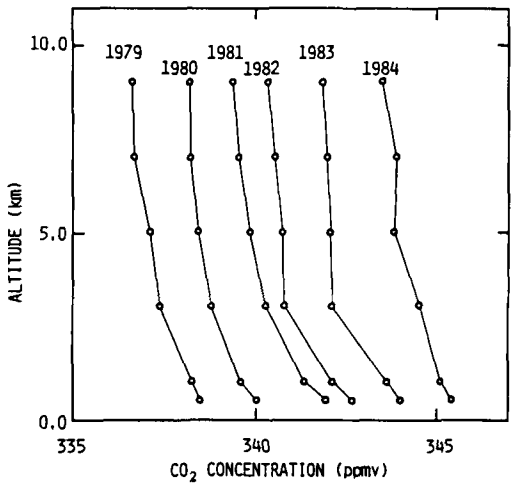

Fig. 5. Vertical profiles of annual mean concentrations of atmospheric CO_2 over Japan, obtained from the best fit curves to measured values.

The vertical profile of the annual mean value of the CO_2 concentration was almost the same from year-to-year, as shown in Fig. 5; the CO_2 concentration decreased gradually with height and the concentration difference between the lowest- and the upper-most layers of the troposphere amounted to about 2 ppmv as an average. Such a mean vertical CO_2 profile, which has also been observed over Scandinavia (Bischof, 1962), is in contrast to that in the southern hemisphere (Pearman and Garratt, 1973; Pearman and Beardsmore, 1984), and implies that the ground surface of northern latitudes acts as a net source of atmospheric CO_2.

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