BALANCE AND BEHAVIOR OF CARBON DIOXIDE AT AN URBAN FOREST INFERRED FROM THE ISOTOPIC AND METEOROLOGICAL APPROACHES

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ABSTRACT. Diurnal variations in $\delta^{14}C$, $\delta^{13}C$ and the concentration of atmospheric carbon dioxide in an urban forest were measured on 9 February 1999 to discriminate and quantify contributions from different CO_2 sources. The biogenic CO_2 concentration remained relatively constant throughout the day. However, anthropogenic CO_2 concentration fluctuated with the atmospheric CO_2 concentration, and seemed to be controlled by wind velocity and the amount of exhaust gases from fossil fuel burning. The vertical profiles of anthropogenic, biogenic, and total CO_2 showed a constant concentration within forest during daytime because of the large vertical CO_2 influx, strong winds, and neutral atmospheric condition. The biogenic contribution at night decreased from the forest floor upwards with a smooth gradient, while the anthropogenic contribution showed a direct mirror because of the location of respective CO_2 sources—the vertical gradient of wind velocity and the horizontal CO_2 supply.

INTRODUCTION

Carbon dioxide emissions due to the burning of fossil fuels such as coal and petroleum increase atmospheric CO_2 concentration and exert strong influence to the greenhouse effect and CO_2 supply to an urban forest. Other human activities such as increasing nutrient status and enhanced temperature (heat island effect) also have a strong influence on urban forests. Hence, understanding the balance and behavior of gaseous CO_2 fluxes within an urban forest will help predict how terrestrial ecosystems may respond to global environmental changes. Here we estimate the CO_2 contributions from anthropogenic and biogenic sources and determined their vertical profiles in order to understand carbon cycling and storage within the urban forest.

The measurements were carried out in winter, when photosynthesis is low, because photosynthesis uses soil respired CO_2, i.e. CO_2 recycling disturbs simple physical mixing relationship (Sternberg 1989). Analyses of radioactive and stable carbon isotopes can identify CO_2 contributions from different reservoirs (Mook 1980; Zondervan and Meijer 1996). Diurnal variations in the CO_2 concentration, $\delta^{14}C$ and $\delta^{13}C$ of the urban atmosphere were measured at two levels—just above the forest canopy and at further higher elevation, to compare the relative contributions of each CO_2 source. In order to understand carbon supply to the urban forest, the diurnal variations of vertical profiles in the CO_2 concentration and $\delta^{13}C$ of the urban forest atmosphere were measured at 6 heights within the forest.

STUDY SITES AND METHODS

The study sites are located about 8 km east of the center of Nagoya City, Aichi Prefecture in Central Japan. Measurements were made from a tower (35°8′57″N, 136°58′28″E) situated in a secondary deciduous forest growing on a gentle slope and a broadcasting tower (Chukyo TV broadcasting Co. Ltd., 35°8′37″N, 136°58′15″E). These two sites are referred to as the forest site and the Chukyo TV

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site, respectively. The forest site is located 600 m east-northeast of the Chukyo TV site. Both tower bases are 66 m above the sea level. The sampling ports were set at 23.75, 19.00, 14.75, 9.10, 5.60 and 1.55 m at the forest site, and at 85 m at the Chukyo TV site. The top canopy is about 12–18 m, although the study was carried out during the leafless period for the oak trees, which are dominant species in the forest. On the tower of the site meteorological instruments were installed. Ten-minute mean values of wind speed, air temperature, relative humidity, 10 Hz fluctuations in the horizontal and vertical wind speed components, and air temperature were measured. Atmospheric stability can be estimated using the parameter of Obukhov's length (L) to be represented as non-dimensional parameter of z/L (Brutsaert 1982), where z is the height on which the value of L is determined. Positive and negative values of z/L indicate stable and unstable atmospheric conditions, respectively. However, the atmosphere can be defined as having a neutral condition when z/L ranges from −1 to 1. The CO_2 fluxes above the forest canopy were calculated using the eddy correlation technique (Kaimal and Finnigan 1994), using the fluctuations in wind speed and CO_2 concentration obtained from a non-dispersive infrared absorption spectrometer, NDIR (KCO-100). The CO_2 efflux from the soil surface (soil respiration) was measured using a closed chamber system connected to a NDIR (LI-6262). On the Chukyo TV site (85 m), wind speed, air temperature, and relative humidity were obtained with an average of 10 minutes.

Air sampling was started at 0:00 on 9 February 1999, and carried out at interval of 3 hr at night and 2 hr in daytime at the forest site; 6 hr at night and 3 hr in daytime at the Chukyo TV site. The air was collected in a 10 L plastic or a 20 L aluminized plastic bag. The CO_2 concentration was measured with a NDIR (LI-6262) as soon as the air sampling was finished. CO_2 was cryogenically trapped from the sampled air within a few hours after sampling. Graphite targets (Kitagawa et al. 1993) were prepared from a sample and a standard (NIST Oxalic acid, Hox-II) for ^{14}C analysis with a Tandetron accelerator mass spectrometer at the Center for Chronological Research, Nagoya University (Nakamura et al. 1985). Stable carbon isotopic ratios were measured on an isotope-ratio mass spectrometer (Finnigan MAT 252) at the same laboratory. The $\delta^{13}C$ values are represented with respect to the PDB standard (Craig 1957), using the NBS-19 standard. A correction of 0.2‰ for N_2O contamination was applied to the $\delta^{13}C$ calculations (Mook and Jongsma 1987).

Atmospheric CO_2 in this study area is mainly a mixture of CO_2 from the free atmosphere and that emitted from anthropogenic and biogenic sources. A mixing model for the three isotopically distinct gases is given by

$$I_S C_S = I_A C_A + I_F C_F + I_B C_B = I_A C_A + I_F (C_S - C_A) f + I_B (C_S - C_A) (1-f) \quad (1)$$

where I is the carbon isotopic value, i.e. $\delta^{14}C$ or $\delta^{13}C$, C is concentration. The suffix S refers to a specific sample in the present study, A is the background (i.e. free) atmosphere, F is fossil fuels (anthropogenic) component and B is the soil respiration (biogenic) component. The fraction of the fossil fuel component to the total excess CO_2 concentration above the background level (C_S–C_A) is f, where $0 \le f \le 1$. In the present study, ^{14}C isotopic value was represented not by $\Delta^{14}C$ but by $\delta^{14}C$, since the discussion of carbon behavior in the present study was explained by the atmospheric mixing of CO_2 released from each source without isotopic fractionation. The end-member values of respective sources were also represented by $\delta^{14}C$ in the present study. This logic is similar to the analysis by Zondervan and Meijer (1996) that used $^{14}C/^{12}C$ ratio. $\delta^{14}C_F$ is defined as −1000‰ theoretically. In study area, $\delta^{14}C_B$ is given as −30 ± 9‰, $\delta^{13}C_A$ is −7.8 ± 0.1‰, $\delta^{13}C_B$ is −27.3 ± 0.4‰, and $\delta^{13}C_F$ is −32.3 ± 1.0‰ (Takahashi 2001). The value of $\delta^{14}C_A$ is given as 136 ± 3‰ (Levin and Kromer 1997), and C_A value is 370.42ppm (Uhse et al. 1998), which were observed in Germany in the winter season of 1996 and 1997. The $\delta^{14}C_A$ was corrected for the decline during 2 years, i.e.

from 1997 to the observation year in the present study, using the exponential curve reported by Levin and Kromer (1997) to be 124‰. C_A was also corrected using the increasing trend of 1.43ppm/year for 1981–92 at Mauna Loa Station (Conway et al. 1994) to be 373.28ppm. Although the present study area (Japan) is far from Germany, it is possible to use these background values in Equation (1) for the following reason: CO_2 concentration and $\delta^{14}C$ values of the background (free) atmosphere in a specific area, e.g. Japan and Germany, can be explained by the simple physical CO_2 mixing between global background and fossil fuel components, since the $\delta^{14}C$ value of CO_2 derived from fossil fuels, which currently controls the discrepancies in CO_2 concentration and $\delta^{14}C$ in the atmosphere from global background, always shows constant to be −1000‰. Over and above, the background CO_2 concentration shows similar values at both areas (Uhse et al. 1998, NIES-Japan http://www-cger.nies.go.jp/moni/gbm-data.html, accessed on 4 October 2000).

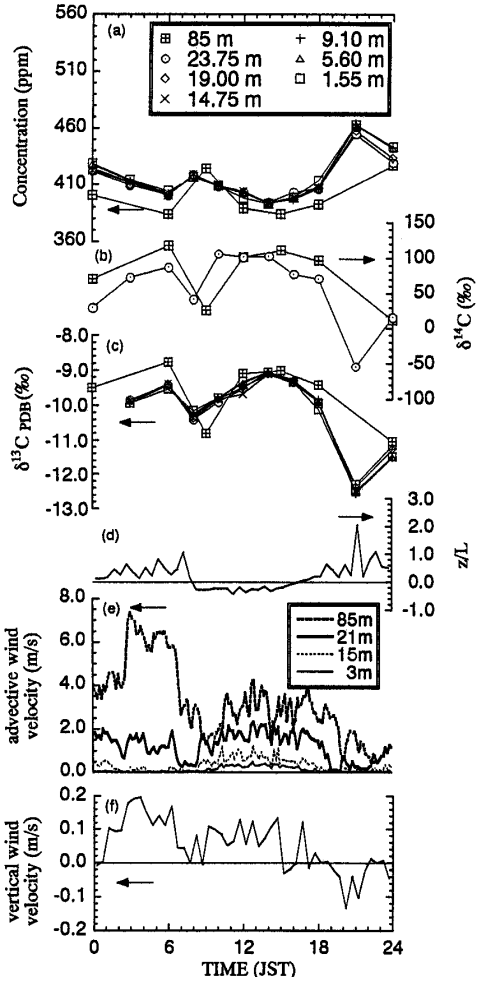

Figure 1 Diurnal variations of concentration (a), $\delta^{14}C$ (b) and $\delta^{13}C$ (c) of the atmospheric CO_2 sampled at 7 heights; 85 m (squared plus), 23.75 m (circle), 19.00 m (diamond), 14.75m (cross), 9.10 m (plus), 5.60 m (triangle) and 1.55 m (square). Also shown are the horizontal wind velocity (d) at 85 m (bold dotted line), 21 m (bold solid line), 15 m (dotted line), and 3 m (solid line), and z/L obtained at 21 m (e).

RESULTS AND DISCUSSION

Variation of Concentration, $\delta^{14}C$ and $\delta^{13}C$ of the Atmospheric CO_2 and Estimate Results

Diurnal variations in the vertical profiles of CO_2 concentration, $\delta^{14}C$, $\delta^{13}C$ and meteorological indices are shown in Figure 1 above (All observed data of CO_2 concentration, $\delta^{14}C$, and $\delta^{13}C$ are listed in the Appendix). CO_2 concentration is generally low during the daytime and high during the night. This tendency was similar to that reported in the previous studies (Aikawa et al. 1995). Diurnal variations in C_F and C_B from the respective sampling sites at 23.75 m and 85 m were estimated from equation 1 using CO_2 concentration and $\delta^{14}C$ (Figure 1a–b), and are shown in Figure 2. The CO_2 concentration of the respective components at 85 m was lower than that at 23.75 m. This stratification is consistent with the fact that CO_2 was released nearby/on the land surface. Large fluctuations were observed for C_F along C_S fluctuations, whereas C_B remained relatively constant throughout the day. This indicated that the fluctuation in CO_2 concentration of the urban atmosphere was mainly explained by anthropogenic CO_2 emission.

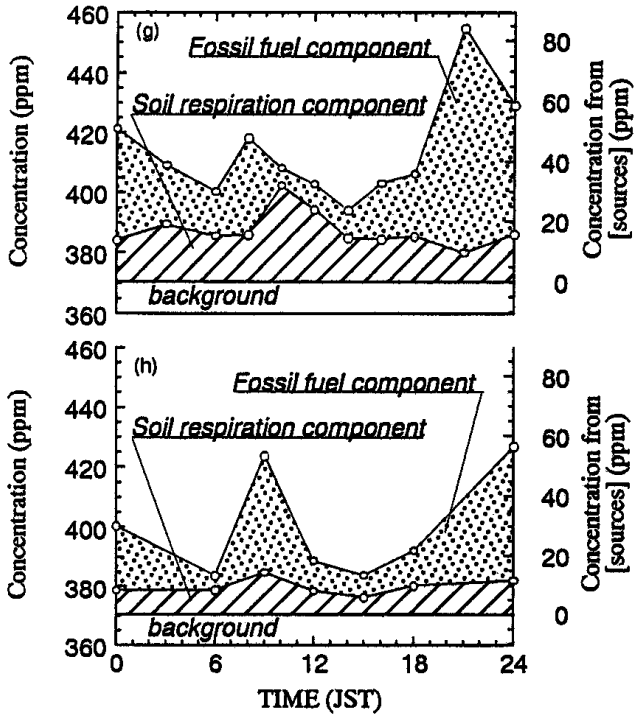

Figure 2 Diurnal variations of CO_2 concentration from the anthropogenic (C_F) and biogenic (C_B) sources at 85 m in the Chukyo TV site (a) and at 23.75 m in the forest site (b). CO_2 concentrations from two sources were shown using different axes.

C_F and the percentage of C_F to C_S (in parentheses) ranged from 2–70 ppm (0.4–15.5%) at 23.75 m, and from 1–41 ppm (0.2–9.5%) at 85 m. These values are very similar to the result from Poland, which was reported to be circa 27 ppm for February 1983–1994 by Kuc and Zimnoch (1998). In the present study area, a large CO_2 emission from automobiles affected the increment of ambient CO_2 concentration. Its influence can not be evaluated in this study, but CO_2 emission from human activities altogether may be considered as the principal contributor to the CO_2 increase (Aikawa et al. 1995). C_B and the percentage of C_B to C_S (in parentheses) ranged from 11–33 ppm (2.4–8.2%) at 23.75 m, and from 7–16 ppm (1.9–3.7%) at 85 m. C_B remained relatively constant throughout the

day, at about 15 ppm (23.75 m) and about 10 ppm (85 m). The constant contribution of C_B is reflected by a constant CO_2 flux from the soil. According to the CO_2 flux measurement using a closed chamber observation in the present study, its diurnal variation being only 0.20 μmol/m^2/sec. with a daily average at the forest site of 0.70 μmol/m^2/sec. This average value of CO_2 flux from soil surface corresponded to 2.7 ppm/hour, if the respired CO_2 was distributed equally within forest atmosphere of 23.75 m height. Thus, mean residence time of soil respired CO_2 might be considered several hours on daily average. We can not assess whether this residence time was reasonable or not, since the rate of air transportation could not be obtained from the meteorological analyses.

Diurnal Variations of CO_2 Concentration and Atmospheric Condition

Air transport from ambient to free atmosphere, which is mainly controlled by the atmospheric stability and wind velocity, causes the diurnal variations of CO_2 concentration. CO_2 concentration in an urban atmosphere does not return to the background level under low wind or stable atmospheric conditions. Atmospheric stability on the observation day in this study was almost under neutral conditions ($-1 \leq z/L \leq 1$) throughout the day (Figure 1e). Hence, the atmospheric stability does not seem to have any influence on the diurnal variations in CO_2 concentration.

We compared CO_2 concentrations of the respective components, i.e. C_S, C_F and C_B, with the wind velocity (Figure 1d). Wind velocity show a negative correlation against both values of C_S ($r = -0.80$) and C_F ($r = -0.76$) with almost identical coefficients. C_B at the Chukyo TV site showed a negative correlation against wind velocity ($r = -0.61$), whereas that at the forest site showed no distinct correlation ($r = 0.28$). The correlation between C_S, C_F and wind velocity indicates that air transportation controls the diurnal variation. On the contrary, C_B showed another trend. At the forest site, the CO_2 from soil respiration was directly supplied from the nearby forest floor, therefore, the diurnal variation of C_B at the forest canopy is not affected by wind velocity at the canopy height. C_B at the forest site increased when a light wind started blowing at the height of 3 m (Figure 1d). Probably, CO_2 from soil respiration at the forest site had accumulated just on the soil surface (e.g. below 0.3 m, Buchmann et al. 1997) until the breeze started. At the Chukyo TV site, the CO_2 from soil respiration was thought to be supplied from the wider urban area. Therefore, it seems that the circulation behavior of CO_2 from soil respiration at a height of 85 m in the atmosphere is almost identical with CO_2 from other components.

Vertical Profiles of Anthropogenic and Biogenic CO_2 within the Forest

Diurnal changes of vertical profiles of the fossil fuel and the soil respiration components within the forest were estimated from equation 1 using CO_2 concentration and $\delta^{13}C$ (Figure 1a, c) that were measured within the forest. The vertical mean profiles of C_F, C_B and C_S during the daytime (8:00 to 16:00) and night (3:00 to 6:00 and 18:00 to 24:00) are shown in Figure 3. Since $\delta^{14}C$ was not measured for all vertical samples, the contributions of the respective components within a forest was estimated using CO_2 concentration and $\delta^{13}C$. Thus, the large errors for the estimated contributions are unavoidable because the $\delta^{13}C$ disparity between the fossil fuel and soil respiration components is small. At night, there is a larger gradient of C_B than C_F, whereas C_B and C_F have constant vertical contributions during the daytime. Diurnal change in gradient of the vertical profile of CO_2 within the forest (Figure 3) is strongly influenced by the wind velocity (Figure 1d). At night, C_B has the decrease trend vertically from the forest floor to the canopy, but C_F has the increase trend. These indicate that CO_2 from soil respiration was directly supplied to the forest atmosphere and CO_2 source of fossil fuel component was not located within the forest. Moreover, the influence of CO_2 from fossil fuel component was stronger at the higher levels in the forest at night. The vertical constant concentration of respective components during the daytime indicates that the forest atmosphere was mixed well vertically.

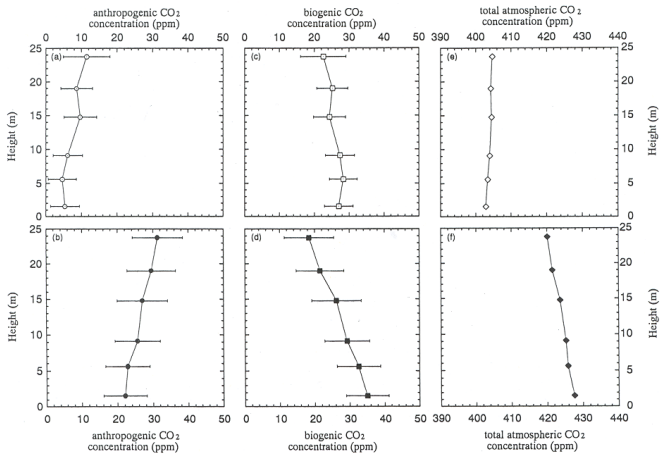

Figure 3 Vertical profiles of the mean CO_2 concentration from anthropogenic (C_F: a, b) and biogenic (C_B: c, d) sources at the daytime (open symbols) and at night (closed symbols). Also shown is the concentration of total atmospheric CO_2 (C_S) at the daytime (e) and at night (f).

We attempted analysis based on meteorological results for CO_2 balance at the forest in order to support the above discussion based on vertical profiles, since the vertical profiles within the forest have a large error caused by the estimation technique. The rate of CO_2 concentration change within the forest atmosphere (ΔQ) can be given by

$$\Delta Q = V + A + R + P \tag{2}$$

where V is vertical CO_2 flux at the forest canopy from outside the forest, A is the advective CO_2 flux from outside the forest horizontally, R is CO_2 evolved by respiration and P is photosynthesis. V was calculated from the fluctuations in wind speed and CO_2 concentration by the eddy correlation technique (Kaimal and Finnigan 1994). The ΔQ value was computed from the diurnal variations of CO_2 concentration profiles. CO_2 flux from soil (R) was measured to be ranged from 2.4 to 3.1 ppm/hr using a chamber styled observation. P is assumed to be negligible. Thus A can be estimated. Diurnal variation of V, A, and ΔQ is shown in Figure 4.

CO_2 was supplied horizontally and escaped vertically at night, whereas the CO_2 movement during daytime showed opposite direction (Figure 4). The constant concentration of fossil fuel and soil respiration components in vertical profiles within the forest during daytime (Figure 3) might be caused by large vertical CO_2 influx and horizontal CO_2 efflux (Figure 4); these exert influence on the vertical air mixing. The strong wind (Figure 1d) and neutral atmospheric condition (Figure 1e) also caused the strong vertical atmospheric mixing within and above the forest. At night, CO_2 was mainly supplied by horizontally (Figure 4). This horizontal influx, i.e. incoming influence, might be stronger at the higher levels in the forest, since the wind velocity show a gradual change vertically (Figure 1d). The smooth vertical profiles within the forest of the respective components at night (Figure 3) also suggested that the horizontal influx of CO_2 to the forest was changed gradually along the height level. The discussion of CO_2 behavior at the urban forest based on CO_2 balance and meteorological results were consistent with analysis from the isotopic approach. Hence, it is possible to consider that the vertical profiles in CO_2 concentration of respective components, C_F and C_B, within the forest (Figure 3) have some specific trends as discussed above, although the estimation error is large.

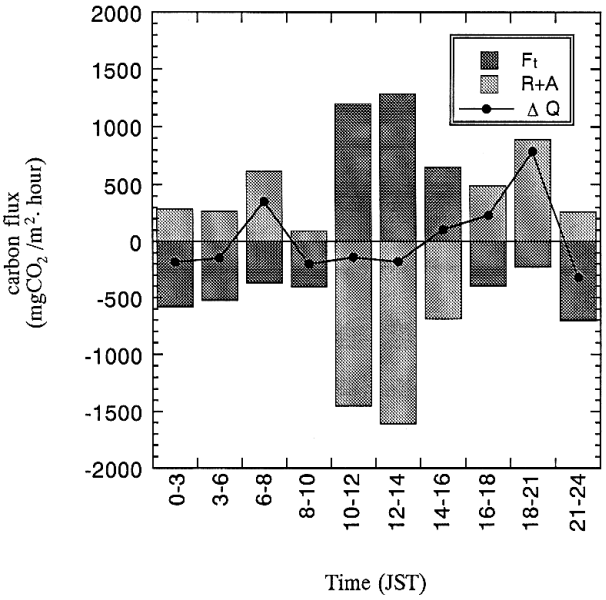

Figure 4 Diurnal variation of vertical CO_2 influx at the top of the forest canopy (V), the advective CO_2 influx from outside the forest (A), and the rate of CO_2 concentration change (ΔQ). The positive and negative values show CO_2 influx and efflux, respectively.

CONCLUSIONS

The diurnal and vertical variations of the anthropogenic and the biogenic contributions in atmospheric CO_2 at an urban area were estimated in winter season using concentration, $\delta^{14}C$ and $\delta^{13}C$ of the atmospheric CO_2. The diurnal variation in the atmospheric CO_2 concentration, which is low during the daytime and high during the night, was mainly controlled by wind velocity with the neutral-condition of atmospheric stability and was influenced by the amount of CO_2 emitted from fossil fuel burning. The contribution of the soil respiration component decreases vertically from the forest floor to the canopy, but that of the fossil fuel component increased. The mean residence time of soil respired CO_2 was suggested to be several hours. The meteorological observation and CO_2 budget analysis at the forest indicate that CO_2 was supplied horizontally and escaped vertically through the forest canopy at night, whereas the CO_2 movement during daytime moved in the opposite direction. These results are consistent with the trends of the vertical profiles in the anthropogenic, biogenic, and total CO_2 concentrations.

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APPENDIX

Table 1 Measured values of concentration, $\delta^{14}C$ and $\delta^{13}C$ in atmospheric CO_2 (height No. 1: 23.75 m; 2: 19.00 m, 3: 14.75 m, 4: 9.10 m, 5: 5.60 m, 6: 1.55 m)

Time-height no.	Concentration (ppm)	$\delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
Forest site			
0–1	421.6	30 ± –7	
0–2	421.6		
0–3	422.5		
0–4	423.9		
0–5	427.7		
0–6	428.4		
3–1	409.5	74 ± –10	–9.55
3–2	409.6		–9.56
3–3	411.4		–9.59
3–4	412.3		–9.64
3–5	413.2		–9.58
3–6	414.0		–9.64
6–1	400.5	88 ± –3	–9.11
6–2	400.6		–9.11
6–3	400.6		–9.09
6–4	401.4		–9.12
6–5	402.5		–9.12
6–6	404.3		–9.23
8–1	418.4	42 ± –9	–10.13
8–2	418.4		–10.10
8–3	418.7		–10.08
8–4	417.7		–10.01
8–5	417.2		–9.96
8–6	416.2		–9.86
10–1	408.5	107 ± –6	–9.61
10–2	408.1		–9.53
10–3	408.8		–9.54
10–4	408.2		–9.50
10–5	408.4		–9.50
10–6	408.9		–9.50

Table 1 Measured values of concentration, $\delta^{14}C$ and $\delta^{13}C$ in atmospheric CO_2 (height No. 1: 23.75 m; 2: 19.00 m, 3: 14.75 m, 4: 9.10 m, 5: 5.60 m, 6: 1.55 m)

Time-height no.	Concentration (ppm)	$\delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
12–1	403.0	102 ± –10	–9.23
12–2	402.5		–9.18
12–3	403.9		–9.38
12–4	403.0		–9.23
12–5	402.7		–9.08
12–6	397.9		–9.02
14–1	394.2	104 ± –11	
14–2	393.6		–8.81
14–3	393.6		–8.81
14–4	394.1		–8.75
14–5	393.6		–8.74
14–6	392.9		–8.76
16–1	403.3	78 ± –12	
16–2	398.8		–9.08
16–3	397.7		–9.02
16–4	396.8		–8.98
16–5	397.2		–8.97
16–6	398.5		–9.05
18–1	406.5	72 ± –8	–9.65
18–2	406.0		–9.60
18–3	407.0		–9.60
18–4	406.6		–9.55
18–5	408.5		–9.61
18–6	413.6		–9.83
21–1	454.8	–54 ± –9	–12.04
21–2	457.7		–12.13
21–3	461.4		–12.25
21–4	463.6		–12.30
21– 5	463.2		–12.27
21– 6	463.1		–12.24
24–1	429.3	17 ± –8	–10.88
24–2	433.5		–10.99
24–3			

Table 1 Measured values of concentration, $\delta^{14}C$ and $\delta^{13}C$ in atmospheric CO_2 (height No. 1: 23.75 m; 2: 19.00 m, 3: 14.75 m, 4: 9.10 m, 5: 5.60 m, 6: 1.55 m)

Time-height no.	Concentration (ppm)	$\delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
24–4	442.6		–11.24
24–5	442.0		–11.23
24–6	443.2		–11.22
Chukyo TV site (85 m)			
0	400.6	71 ± –6	–9.20
6	383.8	118 ± –6	–8.47
9	423.9	26 ± –6	–10.53
12	389.0	103 ± –6	–8.79
15	383.9	111 ± –7	–8.71
18	392.4	97 ± 9	–9.12
24	427.1	13 ± 7	–10.77