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Diurnal and seasonal variations in atmospheric CO_2 in Sapporo, Japan: Anthropogenic sources and biogenic sinks

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Measurements of atmospheric CO_2 were made continuously at a height of 34 m in Sapporo (lat 43.1°N, long 141.3°E), located in northern Japan, from November 2004 to December 2005. Air samples for measurements of atmospheric CO, CH_4, and CO_2 were also taken in Sapporo and Ishikari Hama (lat 43.3°N, long 141.4°E), facing the Japan Sea, at 10- to 14-day intervals during the same period. In Sapporo, the atmospheric CO_2 data showed clear diurnal and seasonal variations. During the diurnal variation, the maximum CO_2 concentration occurred in the morning and the broad minimum in the daytime. The daily mean value of atmospheric CO_2 in the daytime (11–16 JST) was at a maximum in winter to spring and a minimum in summer. From December to February, the daytime atmospheric CO_2 concentration was about 8–13 ppm higher than that in the background air at Ishikari Hama; from July to August, it was nearly equal to that of the background air. In winter, the atmospheric CO concentrations in Ishikari Hama and Sapporo showed a good correlation with atmospheric CO_2 (10 ppb CO/ppm CO_2, $r = 0.82$). However, atmospheric CH_4 remained fairly constant against CO_2 change. In summer, CO_2 flux by the photosynthesis of terrestrial vegetation between 11 and 16 JST was nearly equal to that of area-averaged CO_2 emission (between Ishikari Hama and Sapporo) due to human activities, which was estimated to be 11 μmol $m^{-2}s^{-1}$ in the catchment area.

Keywords: atmospheric CO_2, anthropogenic source, emission ratio, biogenic sink, atmospheric CH_4 and CO

INTRODUCTION

Atmospheric carbon dioxide (CO_2) is a greenhouse gas, the concentrations of which have been increasing due to human activities such as the burning of fossil fuels and deforestation (IPCC, 2001). In urban areas, huge amounts of fossil fuels are consumed by the large numbers of people living there. In order to account for the role of sources for atmospheric CO_2 and to partition anthropogenic CO_2 into various components, studies of atmospheric CO_2 in urban areas have been undertaken (for examples see Grimmond *et al*., 2002, and references cited therein). The atmospheric CO_2 concentrations in urban areas reflect the contributions of both background atmospheric concentrations and variable amounts of CO_2 exchanged with sources and sinks, as shown by the following equation:

$$C_{obs} = C_a + C_s, \quad (1)$$

where C_{obs} is the observed atmospheric CO_2 concentration in an urban area, C_a is the background CO_2 concentration, and C_s is the CO_2 concentration originating from sources and sinks in the catchment area. C_s can be further divided into CO_2 concentrations by anthropogenic (fossil fuel) sources and by photosynthesis and respiration. Previous studies showed higher CO_2 concentrations in urban areas than in the background air. However, factors leading to larger CO_2 concentrations in urban areas vary spatially and temporally. In Nagoya, Japan, the diurnal variation of the anthropogenic CO_2 was the major cause of total atmospheric CO_2 variations, while biogenic CO_2 remained relatively constant throughout the day (Takahashi *et al*., 2002). Kuc *et al*. (2003) reported a reduced consumption of ^{14}C-free fuels, mostly coal, in municipal areas in Krakow and southern Poland after 1991. Clark-Thorne and Yapp (2003) reported the increased consumption of natural gas for home heating and other uses during cooler weather in the Dallas metropolitan area, USA. Pataki *et al*. (2003) showed that the CO_2 concentration and its isotopic composition had a distinct seasonal trend in the Salt Lake valley, USA, due to a com-

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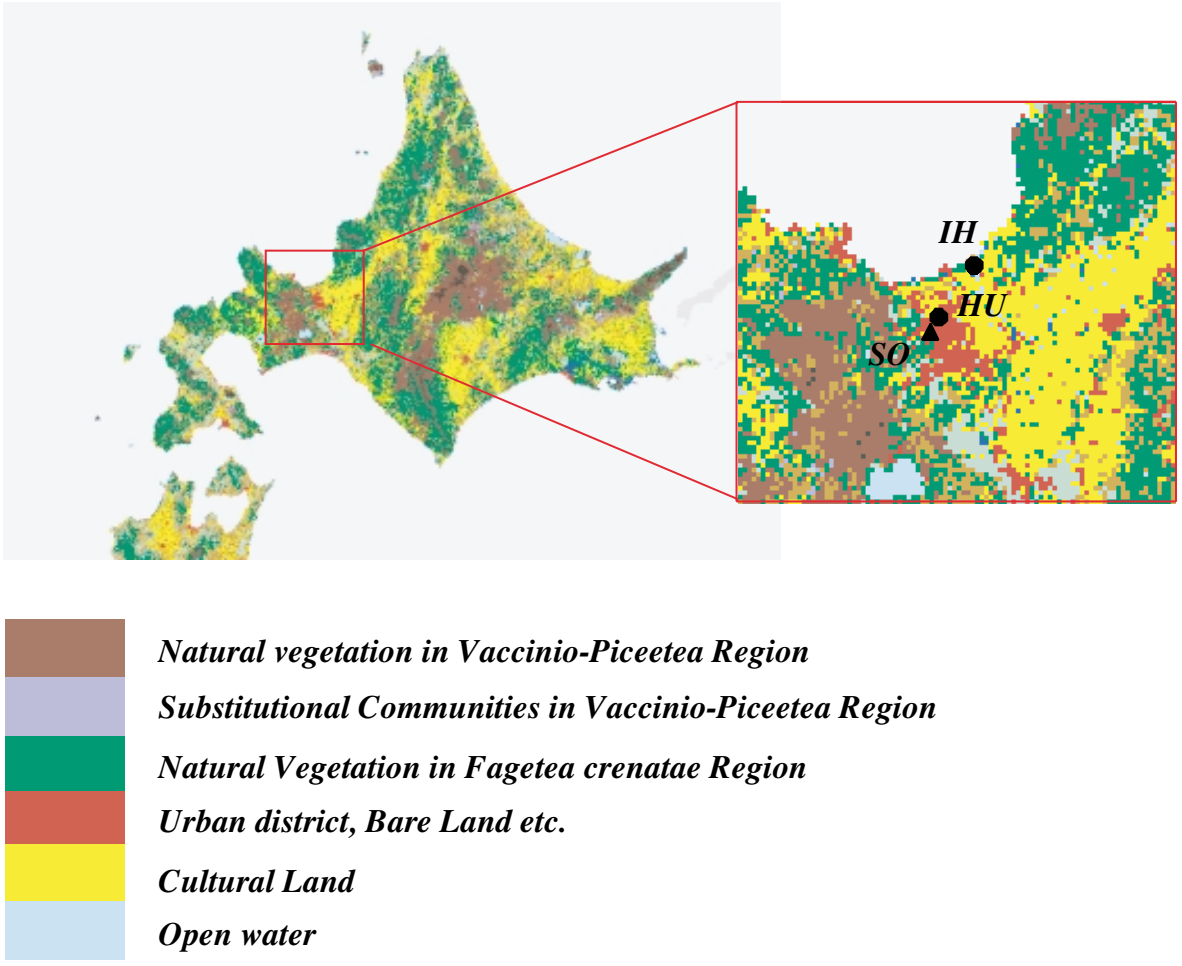

Fig. 1. Location of observation site for measurements of atmospheric CO, CH_4, and CO_2 at the Hokkaido University (HU, solid circle), Ishikari Hama (IH, solid circle), weather observatory in Sapporo (SO, solid triangle), and the actual vegetation map in the northern Japan (http://www.biodic.go.jp/vg_map/vg_html/jp/html/vg_map_frm.html).

bination of varying CO_2 sources and seasonal patterns of atmospheric mixing and transport.

Idso *et al*. (2001) reported that because of urban heat islands and elevated CO_2 concentrations relative to surrounding rural areas, large metropolitan complexes may be valuable analogues of global warming and atmospheric CO_2 enrichment. In addition to elevated temperatures and CO_2 concentrations, other factors, e.g., particulate and photochemical pollutants and urban soils containing heavy metals, affect the urban ecosystem (Gregg *et al*., 2003). Currently, little is known about the net effect of each factor that has a detrimental or beneficial influence on plant growth. In urban areas, it is interesting to examine CO_2 exchange between atmosphere and terrestrial vegetation as well as CO_2 emissions from anthropogenic sources.

In this work, we report diurnal and seasonal variations in atmospheric CO_2 concentrations at a height of 34 m above the ground in the northern area of Sapporo, Japan, from November 2004 to December 2005, as well as relationships of CO and CO_2 with CH_4 and CO_2 in air samples collected in Sapporo and Ishikari Hama, Japan (lat 43.3°N, long 141.4°E), facing the Japan Sea (Fig. 1). Based on these data, CO_2 emissions from anthropogenic sources in winter and CO_2 uptake by terrestrial vegetation in summer are discussed.

Observations

The atmospheric CO_2 concentration was measured at 34 m above the ground on the campus of Hokkaido University (HU) in Sapporo, with a non-dispersive infrared gas (NDIR) analyzer (LI 6262, LI-COR Inc., USA). We installed an air intake at 1 m above the roof of a building on the campus. A diaphragm pump was used to draw air through a 1/4-inch Teflon tube at a rate of 10 L min^{-1}, with most of the air being vented off. Aliquots of sample

Table 1. Meteorological data at the weather observatory in Sapporo from December 2004 to November 2005 (JMA, 2005)

	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
T (°C)	–0.8	–3.5	–3.9	0.1	6.2	10.7	18.3	20.1	23.5	18.8	13.2	5.5
P (mm)	172	153.5	120.5	124	55	62	51	119	114	126	72	150.5
R (MJ/m^2)	4.6	5.8	9.1	12.7	14.2	17.7	20.2	17	17.3	13.1	10.6	6.6
*D (cm)	110	237	139	95	10							4

**D = depth of snow.*

air (0.3 L min^{-1}) were passed through two electric dehumidifiers, Nafion tubing (Perma Pure, Inc., USA), and a chemical desiccant column containing magnesium perchlorate. After the removal of water vapor, the air sample was introduced into a sample cell of the NDIR analyzer, and the output voltage of the NDIR analyzer was integrated at 5-min intervals. The NDIR analyzer was calibrated every 3 hours by successively introducing four calibrated working gases (340, 380, 420, and 450 ppm CO_2 in dry air) into the NDIR analyzer cell for 5 minutes each. The CO_2 concentration of working gas is traceable to the WMO mole fraction scale (Inoue and Matsueda, 2001). Based on replicated measurements of a sample gas in the cylinder, the precision of analysis ($\pm 1\sigma$) was estimated to be better than 0.1 ppm within the range of the CO_2 concentrations of the reference gases.

Concentrations of CO, CH_4, and CO_2 were measured in discrete air samples collected every 10 to 14 days at Ishikari Hama and at the site where continuous measurements of atmospheric CO_2 were made on the HU campus. At Ishikari Hama, air samples were taken around 12 JST under winds from the clean sector, northwest to northeast, and about 1 hour later at HU. At Ishikari Hama, air samples were pressurized manually in 2-L stainless flasks by using a hand-operated diaphragm pump. The apparatus used for the collection of air samples was the same as that reported earlier (Matsueda and Inoue, 1999). At HU, air samples were taken from a manifold used for continuous measurements of atmospheric CO_2. At near-monthly intervals, all flasks were sent to the Meteorological Research Institute, Tsukuba, Japan, for analysis of CO, CH_4, and CO_2. Details of the analytical procedures were reported earlier (Matsueda and Inoue, 1996, 1999).

The campus of HU is located in the northern part of the city of Sapporo. The building in which the Graduate School of Environmental Earth Science is located near the southeast corner of the campus. There is a main street with heavy traffic east of the building (100 m) and a commercial area south of the building (500 m). Important sources of CO_2 are mobile and stationary emissions from fuel combustion.

Sapporo is situated in the southeast corner of the Ishikari plain of Hokkaido (Fig. 1). About 1.85 million people live in the city, which has a total area of 1,121 km^2. The eastern part of Sapporo is a lowland area extending from the Ishikari River to the Nopporo virgin forests. Mountains cover the southern and western parts. Northwest to northeast of Sapporo is the city of Ishikari, population 55 thousand, which covers a total area of 118 km^2. The Ishikari sandbank area to the north faces the Japan Sea. Mountainous forests cover about 60% of the total area of Sapporo and 74% of Ishikari.

Meteorological data collected at the weather observatory in Sapporo (JMA, 2005) from December 2004 to November 2005 are listed in Table 1. From winter to summer, the average temperature increased by more than 25°C. Between December 2004 and March 2005, the ground was continuously covered with approximately 1 m of snow. In winter, winds from the north-northwest prevailed; in summer, winds were from the south-southeast.

Results and Discussion

Atmospheric CO_2, CH_4, and CO

During the winter in Sapporo, the emissions of CO, CH_4, and CO_2 from biogenic sources were expected to be the smallest of the annual cycle because of lower temperatures and a low light intensity. Figure 2 shows the relationships of concentrations of CO and CO_2 (upper panel) to those of CH_4 and CO_2 (lower panel) for discrete air samples collected from December 2004 to February 2005. Atmospheric CO concentrations correlated well with CO_2 concentrations, with a linear regression slope of 10 ppb CO/ppm CO_2 (r = 0.82), which agreed well with the emission ratio in Japan given by Streets *et al.* (2003). Atmospheric CH_4 concentrations remained fairly constant against changes in CO or CO_2 concentrations (0.06 ppb CH_4/ppb CO, r = 0.40; 0.66 ppb CH_4/ppm CO_2, r = 0.39), which deviate widely from those of Street *et al.* (2003): 0.3 ppb CH_4/ppb CO; 2.6 ppb CH_4/ppm CO_2. High CH_4/CO emission ratios of 0.56–0.76 were observed in anthropogenic polluted air in the southeastern USA (Bakwin *et al.*, 1995) and at Mace Head, Ireland

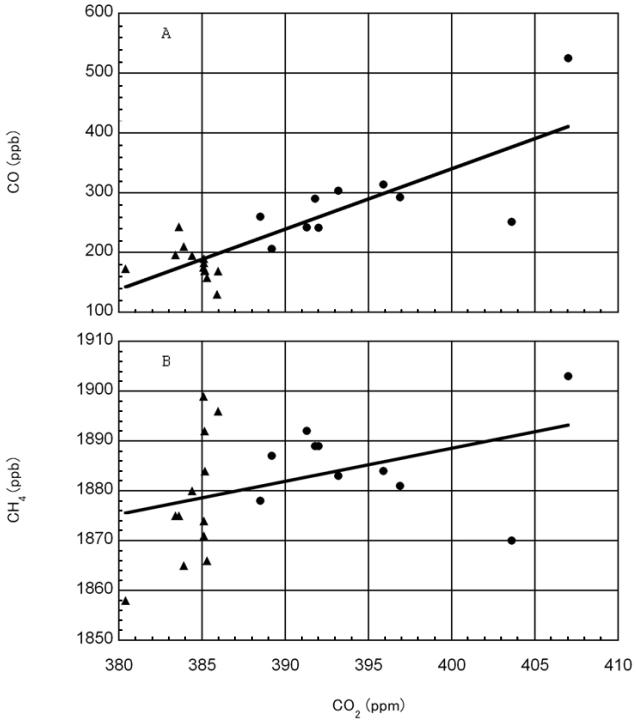

Fig. 2. Scatter plots of CO against CO_2 (upper panel) and CH_4 against CO_2 (lower panel) for air samples collected at Ishikari Hama and HU from December to February. The straight lines show the linear regressions of the data. Solid triangles show data at Ishikari Hama and solid circles at HU.

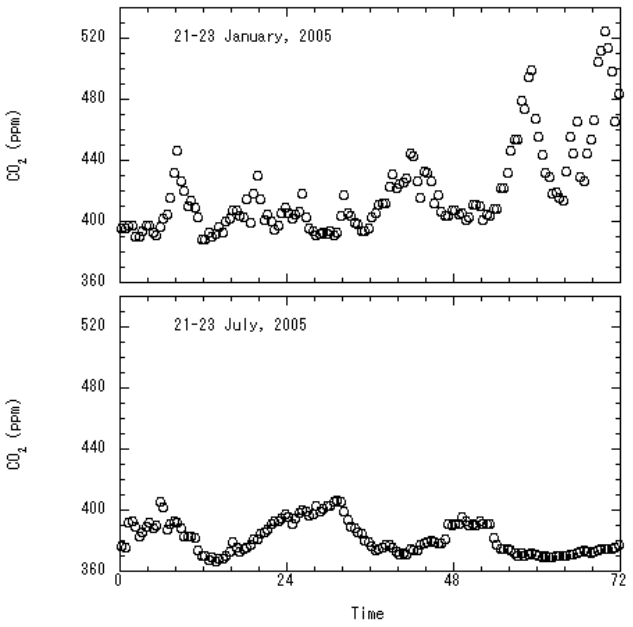

Fig. 3. Diurnal variations in atmospheric CO_2 at HU in Sapporo in 21–23 January 2005 and 21–23 July 2005. Time means elapsed time (hr) since the beginning of 21 January and 21 July.

(McGovern *et al*., 1996). Using estimates of CO_2 emissions for source regions and the measured CH_4/CO_2 ratio in March 1989 (13.5 ppb CH_4/ppm CO_2), Conway *et al*. (1993) estimated a regional European CH_4 source that may be associated with fossil fuel combustion.

During winter in Sapporo, the atmospheric CH_4/CO ratio was within the range of biomass burning, reported to be 0.05 to 0.107 (Matsueda and Inoue, 1999); however biomass burning that could affect the atmospheric CO, CH_4, and CO_2 concentrations has never been reported in and around Sapporo. Present data show that the emissions of CH_4 from anthropogenic sources in Sapporo in winter are low compared with those of the average of Japan, Europe, and the USA. It would be interesting to examine the low wintertime correlation of CH_4 and CO with CH_4 and CO_2 in Sapporo based on emission data from anthropogenic sources; however, such an investigation is beyond the scope of this report.

Diurnal variation

Atmospheric CO_2 measured at 5-min intervals exhibited clear diurnal variations with an occasionally large scatter on time scale of a few ten minutes. Hereafter, we used 30-min averages (Idso *et al*., 2002) in order to obtain the average features of sources and sinks in/around Sapporo. Figure 3 illustrates the atmospheric CO_2 concentration over three days in January and July 2005, which showed the same pattern as that of monthly mean in each month. In January, two maxima occurred: in the morning and at night (8 JST and 20 JST). The atmospheric CO_2 concentration was relatively low a few hours prior to the morning maximum, probably due to decreases in CO_2 emissions and in the afternoon due to convective mixing. Compared with the CO_2 concentration in 21–22 January, the atmospheric CO_2 concentration was extremely high on 23 January. The temperature data measured on the TV tower in the central part of Sapporo showed that the temperature inversion remained below 120 m till 10 JST and developed again 22 JST on 23 January.

In summer, the maximum CO_2 concentration occurred in the morning and the minimum in the afternoon. Over vegetated areas during the plant growth season, the CO_2 concentration reached a maximum near dawn due to the accumulation of respired air and the onset of photosynthesis and solar-induced convective mixing (for examples see Spittlehouse and Ripley, 1977). In Sapporo, the maximum concentration was probably caused by CO_2 emissions due to heavy traffic in the morning as well the above-mentioned effects. The minimum was caused by CO_2 uptake due to photosynthesis and convective mixing.

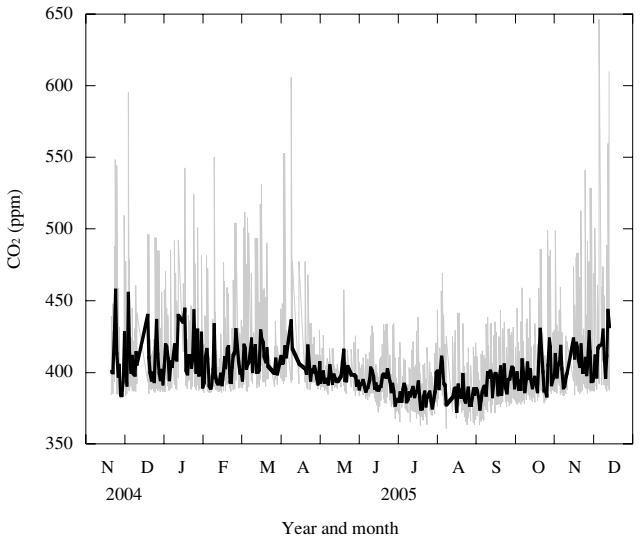

Fig. 4. The daily mean of atmospheric CO_2 concentration (solid line) at 34 m above the ground at Hokkaido University in Sapporo from November 2004 and December 2005. The shaded area encompasses the daily minimum and maximum.

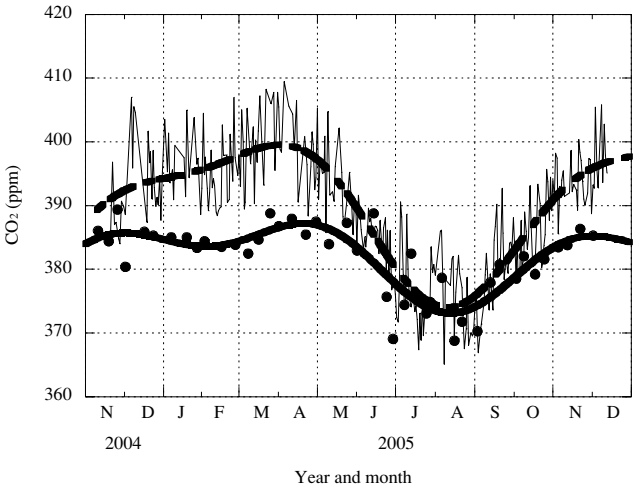

Fig. 5. Seasonal variations in atmospheric CO_2 observed at Ishikari Hama and HU over the period from November 2004 to December 2005. The thick solid curve is the result of curve fitting at Ishikari Hama and thick dashed curve at HU (see text). Solid circle is CO_2 concentration of discrete sir sample taken at Ishikari Hama, and thin solid line the average CO_2 concentration observed at HU between 11 and 16 JST.

Seasonal variations

Figure 4 shows the seasonal variations in daily mean, maximum and minimum concentrations. The maximum amplitude of diurnal variation occurred in winter; the minimum occurred in summer. These variations were caused mainly by the large seasonal variations in the daily maximum concentrations compared with those in the daily minimums.

In order to obtain a representative value of atmospheric CO_2 concentrations in well-mixed air, Thoning *et al*. (1989) conducted data selection based on the variability of hour-to-hour differences in hourly averages of atmospheric CO_2. In Sapporo, sources of atmospheric CO_2 did not allow us to conduct the same data selection as Thoning *et al*. (1989) had done. However, it was possible to obtain CO_2 values in relatively well-mixed air. In this study, we first calculated daily means measured between 11 and 16 JST based on diurnal variations (Subsection "Diurnal variation"). In winter, the average atmospheric CO_2 concentration during the daytime was 416 ppm with a standard deviation (1-σ) of 12 ppm for southerly winds, and 396 ppm with 1-σ of 7 ppm for northerly winds. This result suggests a south-north gradient of atmospheric CO_2 concentrations in winter. In summer, the atmospheric CO_2 concentration during the daytime was 379 ppm with 1-σ of 7 ppm for southerly winds and 374 ppm with 1-σ of 4 ppm for northerly winds, suggesting a fairly constant atmospheric CO_2 concentration over Sapporo.

We fitted daily mean data to a function with a linear long-term trend and two harmonics (1 year and 1/2 year) of Fourier function,

$$xCO_2(t) = \sum_{i=0}^{1} A_i t^i + \sum_{j=1}^{2} \left[B_j \cos(2\pi j t) + C_j \sin(2\pi j t) \right], \qquad (2)$$

where xCO_2 denotes the atmospheric CO_2 concentration and the t the elapsed time in years since November 1, 2004.

The calculated result showed a broad maximum in winter to spring and a minimum in summer (Fig. 5). The peak-to-trough amplitude of the seasonal variation at HU was about 26 ppm, which was greater than that of the background air in the same latitudinal zones (GLOBALVIEW-CO_2, 2006). The daily mean CO_2 concentration was about 8–13 ppm higher than that of the Ishikari Hama from December to February and was almost the same in July and August.

In order to calculate and compare changes in CO_2 concentrations in the surface air at the two sites, it was necessary to know the CO_2 flux between atmosphere and sources or sinks and changes in CO_2 concentrations due to vertical mixing or lateral transport. In this study, for simplicity, we tried to estimate the CO_2 change due to CO_2 emissions from anthropogenic sources. By considering a box of air with a square of side l and some mixing depth H ventilated by a constant horizontal wind speed u, the change of atmospheric CO_2 concentration in the box can be described as follows (Jacob, 1999):

$$\Delta C = C_{obs} - C_a = \left(j_f + j_b \right) \cdot l / (u \cdot H), \qquad (3)$$

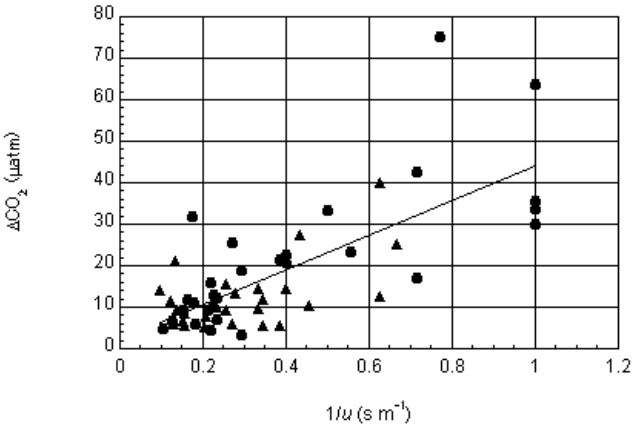

Fig. 6. Changes in CO_2 concentration between Ishikari Hama and HU against the reciprocal of wind speed in January and February 2005. Solid circle is CO_2 concentration in January 2005 and solid triangle in February 2005. CO_2 concentration and wind speed observed at 15 JST were used. The solid line shows the linear relationship: $\Delta CO_2 = 41.54/u + 2.4$ ($r = 0.727$).

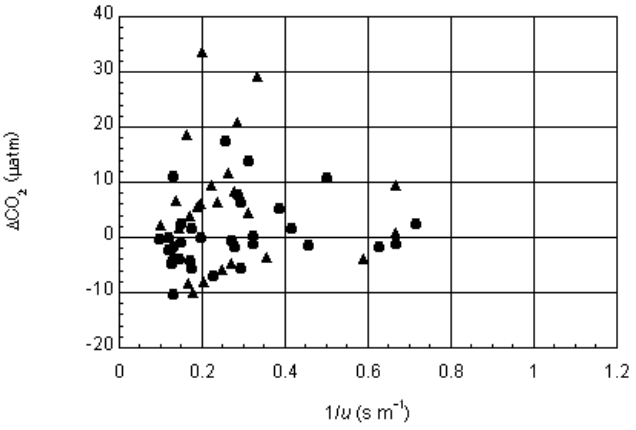

Fig. 7. Changes in CO_2 concentration between Ishikari Hama and HU against the reciprocal of wind speed in July and August 2005. Solid circle is CO_2 concentration and solid triangle in August 2005. CO_2 concentration and wind speed observed at 15 JST were used.

where j_f and j_b are, respectively, the CO_2 flux from anthropogenic CO_2 sources and biological sources/sinks. As mentioned above, in winter, j_b was considered negligibly low compared with j_f. In January and February, increases of CO_2 concentration between Ishikari Hama and HU correlated well with the reciprocal of wind speed (Fig. 6), which allow us to evaluate $j_f l/H$ in Eq. (3). By dividing Japan into boxes with 0.083° by 0.125° (*ca.* 10 km × 10 km), Kannari *et al*. (2004) reported monthly CO_2 emission during the daytime (7–19 JST) and nighttime (19–7 JST) in 1998. During the daytime in 1998, CO_2 emission was estimated to be 28 μmole $m^{-2}s^{-1}$ for the box including HU, 1 μmole $m^{-2}s^{-1}$ for the box including Ishikari Hama. It is clear that CO_2 emission by anthropogenic sources varied largely in and around Sapporo city. By using the average CO_2 emission of three boxes between Ishikari Hama and HU (16 μmole $m^{-2}s^{-1}$) and assuming the increase of CO_2 emission proportional to the per-capita use of fossil fuels between 1998 and 2005 and l of 20 km (distance between Ishikari Hama and HU), the height of H was calculated to be 200 m, which possibly corresponded to the height of atmospheric surface layer in Sapporo.

In contrast to the relationship between CO_2 concentration and the reciprocal of wind speed in January and February, in July and August the atmospheric CO_2 concentration at HU was not correlated with the reciprocal of wind speed (Fig. 7). This was probably caused by the large day-to-day variation (Wang *et al*., 2004) in net ecosystem exchange (NEE). The atmospheric CO_2 concentration at HU was sometimes lower than that of Ishikari Hama due to the larger uptake of CO_2 by photosynthetic activity than the emission of CO_2 from anthropogenic sources.

As mentioned above, in July and August, the CO_2 concentration at HU between 11 and 16 JST was nearly equal to that of Ishikari Hama (Fig. 5). In this case, the mean value of j_b between 11 and 16 JST is expected to be equal to that of $-j_f$. Natural vegetation and cultural land between Ishikari Hama and HU (Fig. 1) assimilated CO_2 at a rate equal to that of CO_2 emission by the anthropogenic sources. In July and August 2005, the area-averaged CO_2 emission during the daytime was estimated to be 11 μmole $m^{-2}s^{-1}$ based on the assumption described above. Because of widely variable anthropogenic sources in space (Kannari *et al*., 2004), the estimation of j_f depends significantly on the wind direction. In the case of south-southeast wind in summer, larger anthropogenic emission in the central part of Sapporo and uptake by terrestrial vegetation in mountainous forest (Fig. 1) occurred, which resulted in relatively constant atmospheric CO_2 concentration at HU during the daytime against changes in wind direction.

At the moment, we do not know the NEE in July/August 2005. According to Wang *et al*. (2004), between 11 and 16 JST in July/August 2001, the NEE of a larch forest in Hokkaido was ranged from 7 to 19 μmole $m^{-2}s^{-1}$ with a mean value of 13 μmole $m^{-2}s^{-1}$ (data are read from figure 3 of Wang *et al*., 2004). In July and August 2005, Kitamura *et al*. (2006) reported a decrease in maximum NEE (18–25 μmole $m^{-2}s^{-1}$) at the site of the FFPRI FluxNet (http://www.ffpri.affrc.go.jp/labs/flux/sap/sape.htm) in the suburbs of Sapporo by 40% as compared with that of 2004, due to the damage by a typhoon which

hit Hokkaido in September 2004. The present method gives us useful information about the average CO_2 flux between air and terrestrial vegetation in rural areas in summer which is difficult to determine using the micro-meteorological technique (Grimmond *et al*., 2002). However, in order to more quantitatively examine from where and to what extent CO_2 exchange between the air and terrestrial vegetation affects atmospheric CO_2 concentration in Sapporo, it is necessary to develop an integrated observation system of measurements by taking into account the atmospheric processes in the catchment area.

SUMMARY

From November 2004 to December 2005, the atmospheric CO_2 concentration was measured at 34 m above the ground on the campus of Hokkaido University (lat 43.1°N, long 141.3°E) in Sapporo, located in northern Japan. During the same period, concentrations of atmospheric CO, CH_4, and CO_2 were also measured in discrete air samples collected every 10 to 14 days in Sapporo and Ishikari Hama (lat 43.3°N, long 141.4°E), facing the Japan Sea. In winter, the atmospheric CO concentrations in Ishikari Hama and Sapporo showed a good correlation with atmospheric CO_2 (10 ppb CO/ppm CO_2, r = 0.82), which agreed well with the emission ratio from the anthropogenic sources in Japan (Streets *et al*., 2003). However, atmospheric CH_4 concentration remained fairly constant against CO_2 change (0.66 ppb CH_4/ppm CO_2, r = 0.39), suggesting that the emissions of CH_4 from anthropogenic sources in Sapporo in winter were relatively low compared with those of the average of Japan, Europe, and USA.

In Sapporo, the atmospheric CO_2 data showed clear diurnal and seasonal variations. During the diurnal variation, the maximum CO_2 concentration occurred in the morning and the broad minimum in the daytime. In winter diurnal variation was caused by the variation in mobile and stationary emissions from fuel consumption and meteorological condition: temperature inversion and the mixing of surface air with background air. In summer, in addition to the effects of anthropogenic sources and meteorological condition, photosynthesis and respiration by terrestrial vegetation play an important role in determining the diurnal variation.

The daily mean value of atmospheric CO_2 in daytime (11–16 JST) was at a maximum in winter to spring and a minimum in summer. From December to February, the daytime atmospheric CO_2 concentration was about 11–13 ppm higher than that in the background air in Ishikari Hama; from July to August, it was nearly equal to that of the background air. In winter, the increase in atmospheric CO_2 between Ishikari Hama and Sapporo was correlated with the reciprocal of wind speed. This suggests the relatively constant day-to-day emission of CO_2 from the anthropogenic sources, which varied widely in space (Kannari *et al*., 2004, 1 μmol $m^{-2}s^{-1}$ in Ishikari Hama and 28 μmol $m^{-2}s^{-1}$ at Hokkaido University in Sapporo in 1998). From July to August, CO_2 flux by the photosynthesis of terrestrial vegetation between 11 and 16 JST was nearly equal to that of area-averaged CO_2 emission between Ishikari Hama and Sapporo (11 μmol $m^{-2}s^{-1}$). The present work gives us useful information about average CO_2 flux between air and terrestrial vegetation in rural area in summer which was reported to be difficult to apply the micro-meteorological technique (Grimmond *et al*., 2002).

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