Title of this paper
A comparison of N_2O and CO_2 concentrations and fluxes in the soil profile between a Gray Lowland soil and an Andosol

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Abstract
We measured N_2O and CO_2 fluxes from the soil surface and in the soil through a depth of 0.3 m, and their concentration profiles through a depth of 0.6 m in both a Gray Lowland soil with macropores and cracks and an Andosol with undeveloped soil structure in central Hokkaido, Japan. The objective of this study was to elucidate the difference of N_2O production and flux in the soil profile between these two soil types. In the Gray Lowland soil, the N_2O concentration above 0.4 m increased with an increase in soil depth. In the Andosol, there were no distinctive N_2O concentration gradients in the topsoil when the N_2O flux did not increase. However, the N_2O concentration at a depth of 0.1 m significantly increased and this concentration was higher than the concentration below 0.2 m when the N_2O flux greatly increased. The N_2O concentration profiles were thus different between these two soils. The contribution ratios of the N_2O produced in the top soil (0-0.3 m depth) to the total N_2O emitted from the soil to the atmosphere in the Gray Lowland soil and the Andosol were 0.86 and 1.00, respectively. It indicates that the N_2O emitted from the soil to the atmosphere was mainly produced in the top soil. However, the contribution ratio of the subsoil to the N_2O emitted from the Gray Lowland soil was higher than that of the Andosol. There was a significant positive correlation between the N_2O flux through a 0.3 m depth and that flux from the soil to the atmosphere in only the Gray Lowland soil. These results suggest that N_2O production in the subsoil of the Gray Lowland soil could have been activated by NO_3^- leaching through macropores and cracks, and subsequently the N_2O produced in the subsoil could have been rapidly emitted to the atmosphere through the macropores and cracks.
Key words: Andosol, carbon dioxide, gas concentration in soil profile, Gray Lowland soil, nitrous oxide

INTRODUCTION

Increased nitrous oxide (N_2O) concentrations in the troposphere cause global warming and contribute to the depletion of the stratospheric ozone (Prather *et al.* 2001). Enhanced N_2O emissions from agricultural and natural ecosystems are believed to be results mainly from the increased use of nitrogen (N) fertilizer. The contribution of agricultural activities to global N_2O emission is estimated to be about 35% (FAO & IFA 2001, Prather et al. 2001). Therefore, elucidation of the mechanism of N_2O emission from agricultural soil has been crucial to mitigate global N_2O emission. Nitrification and denitrification by soil microbes are the dominant processes in the production of N_2O in soils. These processes are strongly influenced by soil conditions such as temperature, water content, nitrate (NO_3^-) and ammonium (NH_4^+) concentrations, and organic matter content (FAO & IFA 2001). Marked increases in N_2O emission rates have been observed immediately after the application of fertilizer and manure (Akiyama et al. 2000, Akiyama & Tsuruta 2002, Jambert et al. 1997, Lessard et al. 1996, Skiba et al. 1996). In addition, some studies reported that the high N_2O emission rates were observed after heavy rain and irrigation (Koga et al. 2004, Kusa et al. 2002, Kusa et al. 2006, Lessard et al. 1996, Mosier & Hutchinson 1981), suggesting that N_2O emissions from the soil to the atmosphere were influenced strongly by N dynamics and the addition of water to soils. In a prismatic structured soil with interstitial pores, water

moves vertically through macropores, bypassing the soil matrix within peds (Hasegawa 1986, Hayashi & Hatano 1999, Inoue 1988). Gas movement is primarily associated with macropores (Osozawa 1998). Also, rainwater or snowmelt water is likely to mix with the soil solution in the topsoil and drain directly through the macropores in the subsoil when large drainage takes place in prismatic structured soil (Hayashi & Hatano 1999). However, in an Andosol characterized by the absence of cracks and fissures after drying, rainwater moves mainly by a matrix flow (Hasegawa & Eguchi 2002) and the movement of gas through macropores becomes minor (Osozawa 1998). Therefore, the movement of NO_3^- and the production and movement of N_2O in soils are influenced by the soil structure. The concentration profiles of soil N_2O have been used to estimate the depth of N_2O production in soils (Goodroad & Keeney 1985, Hosen et al. 2000). A comprehension of N_2O movement in the soil profile is necessary to explain the production and emission of N_2O in the soil.

Carbon dioxide (CO_2) is also one of the greenhouse gases produced by the respiration of soil microbes and roots in soils. Soil microbes and roots are distributed mainly in the top soil (Nakamoto 1993, Osozawa 1998). Therefore, most of the CO_2 emitted is produced in the top soil (de Jong & Schappert 1971). Also, comparisons of N_2O and CO_2 concentrations and fluxes among different soils in the soil profile could be useful to elucidate the influence of soil type on the mechanisms of N_2O production and fluxes in the soil profile. In this study, we measured N_2O and CO_2 fluxes from the soil surface, and both N_2O and CO_2 concentrations and fluxes in the soil profiles in a Gray Lowland soil with macropores and an Andosol without macropores, where N_2O emissions during the pluvial period and after heavy rains were higher than those observed immediately after fertilizer application (Kusa *et al.* 2002, Kusa *et al.* 2006). The objective of this study was to elucidate the difference of N_2O production and flux in the soil profile between these two soil types.

MATERIALS AND METHODS

Experimental sites

The experimental sites were a 2.0×10^4 m^2 onion (*Allium cepa* L.) field in Mikasa City (43° 14' N, 141° 50' E) and a 1.8×10^4 m^2 maize (*Zea mays* L.) field at the National Agricultural Research Center for Hokkaido Region in Sapporo City (43° 00' N, 141° 24' E) located in central Hokkaido, Japan. In the onion field, the soil was Humic Gray Lowland soil (Japanese Society of Pedology 2003); the soil texture at a 0–0.48 m depth was silty clay (SiC) and there were interstitial pores in the subsoil (Table 1). Saturated hydraulic conductivity was low at a depth of 0–0.28 m, and was higher at a depth below 0.28 m (Table 1) because of the presence of macropores (Hayashi & Hatano 1999). Subsurface drains were installed at a 0.8–1.0 m depth and the groundwater table was at 0.7–0.8 m depth throughout the year. Chemical fertilizer was applied to the field at a rate of about 300 kg N ha^{-1} at the end of April. Onion seedlings were transplanted at the beginning of May and harvesting was carried out in both early and mid-September (Kusa *et al.* 2002). In the maize field, the soil was Silandic Andosol (Japanese Society of Pedology 2003). The soil texture at a depth of 0–0.30 m was clay loam (CL) which is rich in humus. An impermeable layer lay 1.3 m below ground level, and consequently the groundwater table temporarily rose to near the ground surface level during the

snowmelt period and after heavy rains (Kanazawa *et al*. 1999). Respective saturated hydraulic conductivity at a depth of 0–0.30 m was lower than that at 0.30–0.47 m (Table 1). Composted cattle manure was applied to the field at a rate of 300 kg N ha^{-1} (fresh weight 3.0 kg m^{-2}) every year in mid-May. After furrowing, chemical fertilizer was applied to the rows at a rate of 130 kg N ha^{-1}. The row width was 0.75 m and inter-row width was 0.25 m. Maize was sown in mid-May and harvested at the end of September. The monitoring of gas emission rates and other factors in the maize field were conducted only between the plants in each row (Kusa *et al.* 2006). For 3 years (1998–2000), N_2O and CO_2 fluxes were usually measured every week on the same day during the snow–free season (Gray Lowland soil: from the end of May to October, Andosol: from June to the end of September) and additionally, N_2O fluxes were measured every week during the snow–free season from 1995 to 1997 in the Gray Lowland soil (Table 2, 3).

N_2O and CO_2 concentration in the soil profile

After the polyvinyl chloride pipes (soil-air sampling tubes: the inside diameter was 0.013 m, the outside diameter was 0.016 m) were installed in the soil, silicon stoppers which were threaded with rubber tubes with three-way cocks were connected to the top of the soil-air sampling tubes. The depths of the soil-air sampling tubes installed were 0.05, 0.1, 0.2, 0.3, 0.4, 0.5 and 0.6 m. Twenty soil-air sampling tubes were installed each at a depth of 0.05 and 0.1 m and ten tubes were installed at various depths between 0.2 to 0.6 m. Gas samples of the enclosed atmosphere in the soil-air sampling tubes were withdrawn using a 10 mL syringe; all gas samples from the same depth were transferred into a 1 L Tedlar®Bag and were mixed. The ambient air above the soil surface was also sampled to obtain the concentration at a 0 m depth. The N_2O concentrations in the gas samples were measured using a gas chromatograph equipped with an electron capture detector (GC-14B; Shimadzu Corp., Kyoto, Japan). The CO_2 concentrations were analyzed using an infrared gas analyzer (ZFP-5; Fuji Electric Co., Ltd., Tokyo, Japan).

Measurement of soil physical properties and rainfall

The soil temperature was measured at a depth of 0.1 m using a digital thermometer. Three undisturbed soil samples (0–0.05 m and 0.05–0.1 m) were collected using three 100 mL steel cylinders at each sampling date; the air-filled porosity, water-filled pore space (WFPS) and the relative gas diffusion coefficient (D/D_0) were measured using the method reported by Osozawa (1998).

To obtain the air-filled porosity and gas diffusion coefficient in the soil profile, undisturbed soil samples were collected once using three 100 mL steel cylinders from the Gray Lowland soil in October, 1996 and from the Andosol in May, 1998 (Gray Lowland soil: 0.05–0.1, 0.15–0.2, 0.23–0.28, 0.32–0.37, 0.43–0.48, and 0.54–0.59 m, Andosol: 0.13–0.18, 0.31–0.36, 0.40–0.45, and 0.58–0.63 m). The air-filled porosity and D/D_0 of these samples were measured at a water suction of –0.098 (water saturated), –0.31, –0.98, –3.1, –9.8, and –31 kPa. The D/D_0 of the water saturated sample was assumed as 0. Regression curves were obtained from the relationship of the soil water suction to the D/D_0 (Table 4) and air-filled porosity. Two tensiometers were installed at depths of 0.1, 0.2, 0.3, 0.4, and 0.5 m and the soil water suction was measured at each

sampling date (Hasegawa & Kasubuchi 1988). The changes in the air-filled porosity and the D/D_0 in the soil profile were calculated using the value obtained from the soil water suction and the regression curves.

Rainfall data for the Gray Lowland soil and the Andosol sites were recorded at the Iwamizawa Weather Station (43° 12.6' N, 141° 47.3' E) (Sapporo Distinct Meteorological Observatory 1995-2000) and the National Agricultural Research Center for Hokkaido Region, respectively.

N_2O and CO_2 fluxes in the soil profile and from the soil surface to the atmosphere

N_2O and CO_2 fluxes through a depth of 0.3 m in the soil profile were calculated using the following equation, using Fick's law (gradient method; Granli & Bøckman 1994) as follows:

$$F_{0.3} = D \times \rho \times \frac{dC}{dz} = \left(\frac{D}{D_0}\right) \times D_0 \times \left(\rho \times \frac{C_{0.4} - C_{0.2}}{z} \times \frac{273}{273+T}\right) \quad (1)$$

where $F_{0.3}$ is the gas flux (mg m^{-2} s^{-1}) in the soil through a depth of 0.3 m, D is the gas diffusion coefficient (m^2 s^{-1}), ρ is the gas density ($\rho CO_2 = \rho N_2O = 1.98 \times 10^6$ (mg m^{-3})) , [dC/dz] is the gas concentration gradient (m^2 m^{-3}), D/D_0 is the relative gas diffusion coefficient at a depth of 0.3 m (these values were calculated from the regression curves of the soil water suction – D/D_0 of the Ap horizon at a depth of 0.23-0.28 m in the Gray Lowland soil and at a depth of 0.13-0.18 m in the Andosol, Table 4), D_0 is the N_2O or CO_2-air inter-diffusion coefficient (m^2 s^{-1}), $C_{0.2}$ and $C_{0.4}$ are the gas concentrations at a depth of 0.2 and 0.4 m (m^3 m^{-3}), respectively, z is the distance from 0.4 to 0.2 m, and T is the soil temperature between 0.2 and 0.4 m (°C) which was presumed to be 20 °C. D_0 under the air pressure 1 atm and the soil temperature 20 °C were calculated using the following equation (Pritchard & Currie 1982):

$$D_0 = Ds \times \left(\frac{273+20}{273}\right)^{1.79} \quad (2)$$

where D_S (N_2O) and D_S (CO_2) (m^2 s^{-1}, in standard condition) represent 0.143×10^{-4} and 0.139×10^{-4}, respectively (Pritchard & Currie 1982).

In our previous paper (Kusa *et al.* 2008), we revealed that the gradient method was useful in measuring the N_2O fluxes from the soil surface into the atmosphere (flux from the soil surface). However, there were differences in the CO_2 and extremely high N_2O fluxes between chamber and gradient methods when the production and consumption of these gases were active in the soil above the installed location of the soil-air sampling tube. Therefore, the N_2O and CO_2 fluxes from the soil surface were measured by a closed-chamber method. Cylindrical stainless steel chambers, 0.3 m in diameter and 0.35 m high for the Gray Lowland soil and 0.2 m in diameter and 0.2 m high for the Andosol, were used. Fifteen minutes after placement of the chamber, a gas sample was taken from the enclosed atmosphere. The mean gas emission rates of four replications in the Gray Lowland soil and of two replicates in the Andosol were calculated. The gas sampling method and the calculation of gas fluxes were described in detail in our previous papers (Kusa *et al.* 2002, 2008). The cumulative gas fluxes during the study period were calculated through linear interpolation.

Mass balance analysis

Hosen et al. (2000) showed that N_2O consumption in the top soil (above 0.24 m) does not have much effect on the N_2O emission rate. Although CO_2 can be dissolved in the soil water, Osozawa (1998) reported that the CO_2 runoff volume by water percolation was very small. Therefore, mass balance analyses were conducted to estimate the N_2O and CO_2 productions of topsoil (0–0.3 m) by the following equation:

$$P = F_0 - F_{0.3} + (M_e - M_s)$$

where P is the N_2O and CO_2 production (mg m^{-2}) in the topsoil during the study period, and F_0 and $F_{0.3}$ are the cumulative N_2O and CO_2 fluxes (mg m^{-2}) from the soil surface through a depth of 0.3 m during the study period. M_s and M_e signify the mass of N_2O and CO_2 (mg m^{-2}) respectively, in the topsoil at the beginning and end of the investigation which is the product of air-filled porosity (m^3 m^{-3}), gas concentration (mg m^{-3}), and depth (m). The contribution ratios of the gas production (P/F_0) in the topsoil to the gas emitted from the soil surface to the atmosphere were estimated.

RESULTS

Seasonal patterns of rainfall and soil physical properties

The frequency of rainfall in both Gray Lowland and Andosol sites increased after July every year (Figs. 1 and 2). The mean values of WFPS from 1998 to 2000 at depths of 0–0.05 and 0.05–0.1 m were 45 and 59 % in the Gray Lowland soil and 48 and 57 % in the Andosol, respectively (Figs. 3 and 4). At a depth of 0–0.1 m, the values of D/D_0 were below 0.02 when the values of WFPS were above 60%. The values of D/D_0 in the Andosol were higher than those of the Gray Lowland soil when the values of WFPS were below 60% (Fig. 5). The mean values of soil water suction at depths of 0.2, 0.3, and 0.6 m from 1998 to 2000, were –15.6, –10.9, and –3.2 kPa in the Gray Lowland soil and –10.5, –9.9, and –3.6 kPa in the Andosol. There were no significant differences in WFPS (paired t–test: 0–0.05 m $|t|$=2.02, p = 0.05, n = 61 and 0.05–0.1 m $|t|$=1.07, p=0.29, n=59) and soil water suction (paired t–test: 0.2 m $|t|$=1.82, p = 0.08, n = 24, 0.4 m $|t|$=1.28, p=0.21, n=24 0.6 m $|t|$=0.11, p=0.91, n=21) between the Gray Lowland soil and the Andosol. The soil water suction increased with an increase in soil depth. The value of WFPS increased while the soil water suction decreased after rainfall (Figs. 1–5). Also, the soil temperature at a depth of 0.1 m increased from spring to summer and decreased after summer (Figs. 3 and 4).

In 1998–2000, the mean values of soil air porosity at a depth of 0–0.05, 0.05–0.1, 0.3, and 0.6 m were 32, 23, 5.5, and 4.9 % in the Gray Lowland soil and 35, 29, 8.7, and 7.6 % in the Andosol, respectively. The values of soil air porosity in the Andosol were higher than those of the Gray Lowland soil (paired t–test: depth of 0–0.05 m $|t|$=2.65, p=0.01, n=61, depth of 0.05–0.1 m $|t|$=4.60, p<0.01, n=59, depth of 0.3 m $|t|$=1.84, p=0.08, n=18, depth of 0.6 m $|t|$=3.48, p<0.01, n=21). The mean values of D/D_0 at depths of 0–0.05, 0.05–0.1, 0.3, and 0.6 m (1998–2000) were 0.097, 0.048, 0.003, and 0.003 in the Gray Lowland soil and 0.150, 0.080, 0.004, and 0.003 in the Andosol, respectively (Fig. 6). The values of D/D_0 above 0.3 m in the Andosol were higher than those in the Gray Lowland soil, but there was no significant difference at a depth of 0.6 m (paired t–test: depth of 0–0.05 m $|t|$=4.41, p<0.01, n=61, depth of 0.05–0.1 m $|t|$=5.12,

p<0.01, n=57, depth of 0.3 m $|t|$=2.39, p=0.03, n=18, depth of 0.6 m $|t|$=1.03, p=0.31, n=24).

Concentration and flux of N_2O in the soil profile

The cumulative N_2O flux from the soil to the atmosphere during the study period from 1995 to 2000 ranged from 310 to 1190 mg N m^{-2} in the Gray Lowland soil and that cumulative flux from 1998 to 2000 ranged from 630 to 1980 mg N m^{-2} in the Andosol (Table 2). There was no significant difference in the cumulative N_2O flux between the Gray Lowland soil and the Andosol from 1998–2000 (t-test: $|t|$=1.11, p=0.33, n=3). Also, a significant increase in N_2O fluxes occurred during the increasing frequency of rainfall in the Gray Lowland soil (Fig. 1a) and after heavy rainfall (above 80 mm day^{-1}) in the Andosol (Fig. 2a).

In both the soils, the N_2O concentrations at a depth of 0.05 m were always higher than those of the ambient air, which is about 0.3 ppmv (10^{-6} m^3 m^{-3} = ppmv). A significant increase in soil N_2O concentrations occurred after July in the Gray Lowland soil, when the frequency of rainfall increased and after heavy rainfall occurred (above 80mm day^{-1}) in the Andosol. These increases in concentration were greater than those that took place in June after fertilizer applications (Figs. 1 and 2). The seasonal pattern of the N_2O concentration in the soil was similar to the N_2O flux from the soil to the atmosphere. The mean values of the N_2O concentration in the soil at depths of 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, and 0.6 m were 2.2, 5.8, 15, 22, 54, 62, and 59 ppmv in the Gray Lowland soil and 8.4, 18, 7.7, 7.2, 11, 16, and 10 ppmv in the Andosol, respectively. The maximum concentrations of N_2O at these depths were 21, 37, 83, 140, 240, 430, and 370 ppmv in the Gray Lowland soil, and 93, 250, 55, 18, 59, 110, and 35 ppmv in the Andosol, respectively. In the Gray Lowland soil, the N_2O concentrations above 0.4 m increased with an increase in soil depth, however, there was no increase in concentration below a 0.4 m depth (Fig. 7). Furthermore, the N_2O concentration gradients of the soil profile increased from August to October (Figs. 1 and 7). In the Andosol, there were no N_2O concentration gradients in the topsoil in June when the N_2O flux did not increase. However, the N_2O concentration at a depth of 0.1 m significantly increased (above 40 ppmv), and this concentration was higher than that at a depth of 0.2 m when the N_2O flux greatly increased (September 1998, July 1999, July 2000, and September 2000) (Figs. 2 and 8).

The mean value of N_2O fluxes in the soil through a depth of 0.3 m in the Gray Lowland soil was 0.026 mg N m^{-2} h^{-1} and it was about ten times higher than that in the Andosol (0.002 mg N m^{-2} h^{-1}). The N_2O fluxes in the soil through a depth of 0.3 m were much lower than those from the soil to the atmosphere (Figs. 1 and 2). There was a significant positive correlation between the N_2O flux at a depth of 0.3 m and the N_2O flux from the soil to the atmosphere in the Gray Lowland soil (r= 0.54, p<0.01, n=90). However, there was no significant correlation in the Andosol.

The cumulative N_2O flux in the soil through a depth of 0.3 m during the study period ranged from 56 to 160 mg N m^{-2} (mean value: 87 mg N m^{-2}) in the Gray Lowland soil and from 1.3 to 7.8 mg N m^{-2} (mean value: 5.1 mg N m^{-2}) in the Andosol (Table 2). The cumulative N_2O flux of the Gray Lowland soil was significantly higher than that of the Andosol in 1998 to 2000 (t-test: $|t|$=3.21, p<0.05, n=3). In both the soils, the cumulative N_2O fluxes in the soil through a depth of 0.3 m were lower than those from the soil to

the atmosphere. The N_2O produced in the soil above a depth of 0.3 m during the study period were 240–1030 mg N m^{-2} (mean value: 590 mg N m^{-2}) in the Gray Lowland soil and 640–1980 mg N m^{-2} (mean value: 1350 mg N m^{-2}) in the Andosol (Table 2). The contribution ratios of the N_2O produced in the topsoil (above the depth of 0.3 m) to the emitted N_2O from the soil to the atmosphere were 0.77–0.91 in the Gray Lowland soil and 0.99–1.01 in the Andosol (Table 2). These contribution ratios of the Gray Lowland soil were significantly higher than those of the Andosol from 1998 to 2000 (t-test: $|t|=6.40$, $p < 0.01$, $n = 3$). In other words, the proportion of the N_2O produced in the subsoil (below a depth of 0.3 m) to the N_2O emitted from the soil to the atmosphere was 9–23 % in the Gray Lowland soil and 0–1 % in the Andosol.

Concentration and flux of CO_2 in the soil profile

The cumulative CO_2 fluxes from the soil to the atmosphere in the Gray Lowland soil and the Andosol during the study period were 360–430 g C m^{-2} and 340–540 g C m^{-2}, respectively (Table 3). There was no significant difference in the CO_2 emission from the soil into the atmosphere between the Gray Lowland soil and the Andosol (t-test: $|t|=0.20$, $p=0.85$, $n=3$). In both the soils, the CO_2 flux increased in July and August with an increase in soil temperature (Figs. 3 and 4).

The mean values of the CO_2 concentration in the soil at depths of 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, and 0.6 m were 2.1, 3.5, 7.6, 12, 20, 21, and 21×10^3 ppmv in the Gray Lowland soil and 3.5, 6.4, 10, 13, 17, 22, and 21×10^3 ppmv in the Andosol, respectively. In both the soils, CO_2 concentrations at a depth of 0.05 m were always higher than that of the ambient air (0.36×10^3 ppmv). The CO_2 concentration gradient in the soil profile also increased from July to September with an increase in soil temperature. The seasonal pattern of CO_2 concentration in the soil was similar to that of the CO_2 flux from the soil surface (Figs. 3 and 4). The CO_2 concentration in the soil above a depth of 0.4 m increased with an increase in depth; however, the concentration below a depth of 0.4 m did not increase (Figs. 7 and 8).

The mean value of CO_2 fluxes in the soil through a depth of 0.3 m was 5.5 mg C m^{-2} h^{-1} in the Gray Lowland soil and 2.6 mg C m^{-2} h^{-1} in the Andosol. The CO_2 fluxes in the soil through a depth of 0.3 m were much lower than those from the soil to the atmosphere (Figs. 3 and 4). There was no significant correlation between the CO_2 flux in the soil through a depth of 0.3 m and CO_2 flux from the soil to the atmosphere in both the soils.

The range of the cumulative CO_2 flux in the soil through a depth of 0.3 m during the study period through a depth of 0.3 m was 16–44 g C m^{-2} (mean value 27 g C m^{-2}) in the Gray Lowland soil and 3.7–7.1 g C m^{-2} (mean value 5.2 g C m^{-2}) in the Andosol, and this cumulative CO_2 flux of the Gray Lowland soil was higher than that of the Andosol from 1998 to 2000 (t-test: $|t|=2.51$, $p=0.07$, $n=3$) (Table 3). In both soils, the cumulative CO_2 fluxes in the soil through a depth of 0.3 m were lower than those from the soil to the atmosphere. The CO_2 produced in the soil above a depth of 0.3m during the study period was 330–410 g C m^{-2} (mean value 370 g C m^{-2}) in the Gray Lowland soil and 320–530 g C m^{-2} (mean value 410 g C m^{-2}) in the Andosol. The contribution ratios of the CO_2 produced in the topsoil (above the depth of 0.3 m) to the CO_2 emitted from the soil into the atmosphere were 0.89–0.96 in the Gray Lowland soil and 0.99 in

the Andosol (Table 3). These contribution ratios of the Andosol were significantly higher than that of the Gray Lowland soil from 1998 to 2000 (t-test: $|t|$=2.88, p=0.04, n=3). In other words, the proportions of the CO_2 produced in the subsoil (below the depth of 0.3 m) to the CO_2 emitted from soil to the atmosphere were 4–11% in the Gray Lowland soil and 1% in the Andosol.

DISCUSSION

The N_2O concentration in the soil profile

In both Gray Lowland and Andosol soils, the seasonal pattern of the N_2O flux from the soil surface was similar to that of the N_2O concentration in the soil from depths of 0.05 to 0.6 m (Figs.1 and 2). In the Gray Lowland soil, the N_2O concentration gradient from the surface soil to a depth of 0.6 m increased when the N_2O flux significantly increased (Figs. 1 and 7). On the other hand, the N_2O concentration gradient of the surface soil increased when the N_2O flux significantly increased in the Andosol (Figs. 2 and 8). Therefore, it suggests that the N_2O produced in the soil profile was emitted into the atmosphere. In several studies, a similar N_2O flux from the soil into the atmosphere and a concentration gradient in the soil profile was reported after fertilizer application and irrigation (Mosier & Hutchinson 1981, Goodroad & Keeney 1985, Lessard et al. 1996, Li et al. 2002, Müller et al. 2004, Clough et al. 2006, van Groenigen et al. 2005, Hirose & Tsuruta 1996). In our previous paper, we reported that a significant amount of N_2O emission from the Gray Lowland soil and the Andosol occurring during the increasing frequency of rainfall and after heavy rainfall was derived from the denitrification process (Kusa *et al.* 2002, Kusa *et al.* 2006). In some reports, it was suggested that the effect of soil moisture to the N_2O production by the denitrification process was greater than that of the NO_3^- concentration in the soil. This is because N_2O could be produced by denitrification in the subsoil (below the depth of 0.2 m) with high soil moisture levels and a low NO_3^- concentration (Li et al. 2002, Müller et al. 2004, Van Groenigen et al. 2005). In both the Gray Lowland soil and the Andosol, a significant increase in the N_2O flux with an increase in soil moisture and after heavy rainfall, and with the increasing N_2O concentrations in the top soil and the decreasing soil water suction occurred at the same time (Figs. 1 and 2). These results suggest that denitrification is the main process attributed to the production of N_2O in the soil.

The maximum N_2O concentrations around a depth of 0.1 m were reported to be 0.9–180 ppmv (Mosier & Hutchinson 1981, Goodroad & Keeney 1985, Arah et al. 1991, Lessard et al. 1996, Li et al. 2002, Jacinthe & Lal 2004, Müller et al. 2004, van Groenigen et al. 2005, Itahashi et al. 1998). In our study, the maximum N_2O concentration at a depth of 0.1 m in the Gray Lowland soil was 37 ppmv (this N_2O flux was 1.5 mg N m^{-2} h^{-1}) (Fig. 1), this value remained within the reported maximum N_2O concentrations (Mosier & Hutchinson 1981, Goodroad & Keeney 1985, Arah et al. 1991, Lessard et al. 1996, Li et al. 2002, Jacinthe & Lal 2004, Müller et al. 2004, van Groenigen et al. 2005, Itahashi et al. 1998) and it was similar to the report from a corn field in Colorado (the N_2O concentration at the depth of 0.1 m was about 40 ppmv and the N_2O flux was about 2.3 mg N m^{-2} h^{-1}) (Mosier and Hutchinson 1981). On the other hand, the maximum N_2O concentrations around a depth of 0.1 m were about 0.4–4.2 ppmv in the Japanese Andosols, when maximum N_2O fluxes (0.04–0.2 mg N m^{-2} h^{-1})

were measured just after fertilizer application (Tsuruta 1997, Yoh et al. 1997, Li et al. 2002, Hirose & Tsuruta 1996). The N_2O concentrations in Japanese Andosols were lower than those in other soils (Mosier & Hutchinson 1981, Goodroad & Keeney 1985, Arah et al. 1991, Lessard et al. 1996, Jacinthe & Lal 2004, Müller et al. 2004, van Groenigen et al. 2005), and this result is consistent with the values reported by Akiyama and Tsuruta (2003), who concluded that N_2O emissions from Japanese Andosols were lower than those from other soils in Japan and the world. The reason for low N_2O concentrations in Japanese Andosols was pointed out to be the high gas diffusivity due to high porosity and low N_2O production by denitrification (Li *et al.* 2002). However, the Andosol of our study site showed that the maximum N_2O concentration at a depth of 0.1 m was 250 ppmv and this concentration was higher than the other reported values, especially from Japanese Andosols (Figs. 1 and 2). Therefore, a lot of N_2O could have possibly been emitted from the Japanese Andosols which had high groundwater levels and a high soil moisture level after heavy rainfall (as in our study site). This is because the N_2O concentration in the soil surface might have increased due to denitrification after heavy rain on these soils.

The N_2O concentration profiles in the soils were different between the Gray Lowland soil and the Andosol (Figs. 7 and 8). N_2O concentration profiles of some studies (Mosier & Hutchinson 1981, Arah et al. 1991, Burton & Beauchamp 1994, Yoh et al. 1997, Li et al. 2002, Müller et al. 2004, Jacinthe & Lal 2004, van Groenigen et al. 2005) were similar to the profile in the Gray Lowland soil of our study site, where the concentration in the soil increased in the deeper layer. In Japanese Andosols, it was reported that N_2O concentrations in the soil surface (depth of 0.1 to 0.2 m) were higher than that in the deeper layers (Hirose & Tsuruta 1996, Itahashi et al. 1998, Tsuruta 1997). Although this result is consistent with our study, the N_2O concentration profile in the soil varied according to the seasons and was different among the Japanese Andosols (Yoh et al. 1997, Verchot et al. 1999, Li et al. 2002). In this way, there are no consistent results about the N_2O concentration profiles in the soil.

The CO_2 concentration in the soil profile

In both soil types, the CO_2 concentration in the soil increased from spring to summer and decreased in autumn. The seasonal pattern of this concentration was similar to that of the CO_2 flux from the soil to the atmosphere (Figs. 3 and 4). The CO_2 concentrations at a depth of 0.05 m were always higher than that of the ambient air. Similar types of results have been frequently reported (de Jong & Schappert 1971, Hendry et al. 1999, Jacinthe & Lal 2004, Osozawa 1998). Also, it is reported that the peak of the CO_2 concentration in the soil profile gradually dropped from a depth of 0.2–0.4 m with growing plants and the CO_2 concentration increased with the depth at both fallow and cultivated soils after the autumn season (de Jong & Schappert 1971, Hendry et al. 1999, Jacinthe & Lal 2004, Osozawa 1998). Similar results were confirmed in our study sites (Figs. 3, 4, 7, 8).

N_2O and CO_2 concentrations and fluxes after rainfall

The CO_2 concentration in the soil was greatly influenced by rain. This is because, at first, the CO_2 concentration in the surface soil increased just after rainfall when the soil porosities were filled by rainwater. Thus, the CO_2 concentration in the surface soil could

have been vertically diffused after drainage and evaporation from the soil surface, resulting in a decrease in CO_2 concentrations around the soil surface (Osozawa 1998). In the Andosol, the CO_2 concentration at a 0.1 m depth increased after heavy rainfall (the precipitation during a week before the day of investigation exceeded 80 mm in July 1999, July 2000, and September 2000), but the CO_2 fluxes decreased (Figs. 3, 4). This could possibly be due to the fact that the gas diffusion from the soil into the atmosphere could have been restricted by rainfall, as reported by Osozawa (1998). This is because the D/D_0 of the surface soil at this time was below 0.02 (Figs 3, 4, and 5), which might have restricted gas diffusion from the soil into the atmosphere (Hatano 1997). However, the N_2O concentrations in the surface soil and N_2O flux from the soil surface increased at the same time (Figs. 1, 2). Therefore, it suggests that anaerobic conditions with increasing soil moisture levels and the restriction of gas diffusion could have accelerated the production of N_2O by denitrification, in the surface soil.

The effect of soil structure to production and emission of N_2O

N_2O production in the lower soil profile was reported in several studies when NO_3^- leached from the surface layer after rain (Goodroad & Keeney 1985, Müller et al. 2004, van Groenigen et al. 2005). It suggests that a N_2O production spot in the soil could be greatly influenced by water movement and the NO_3^- concentration in the soil. In the Gray Lowland soil of our study, the NO_3^- in the surface soil leached through macropores after rain (Hayashi & Hatano 1999), and the total N concentration of the groundwater rapidly increased after applications of slurry in the grassland (Kanazawa et al. 1999) adjoining the Andosol site of our study. Therefore, it seemed that the NO_3^- in the surface soil leached with rain water in both soils. In the Gray Lowland soil of our study site, the NO_3^- concentrations of the soil solution at a depth of 0.7 m were always below 3 mg N L^{-1}, while the concentrations of the pipe drain were always around 10 mg N L^{-1} (Hayashi & Hatano 1999), therefore the subsoil around the macropores could have been in contact with high concentrations of NO_3^-. On another front, it was reported that the NO_3^- concentrations in the soil solution at a depth of 0.8 m and those in the seepage water were at the comparable level in the other Andosol in Hokkaido, Japan (Suzuki & Shiga 2004). Hasegawa and Eguchi (2002) reported the rainwater moved mainly by a matrix flow in an Andosol without cracks and fissures in Tsukuba, Japan. Therefore, the NO_3^- concentration in the surface soil could have been higher than in the subsoil, because water and NO_3^- infiltrated from the surface soil to the subsoil by matrix flow. This suggests that the activity of N_2O production in the subsoil of the Gray Lowland soil was higher than that in the Andosol. It corresponded with the difference in N_2O concentration profiles in the soil (Figs 1 and 2) and with the contributing ratio of N_2O production in the top soil between both soils (Table 2).

An important factor regarding N_2O emission from the soil into the atmosphere is that it was not only the activity of N_2O production in the soil, but also the gas diffusivity. Under the usual upland soil moisture conditions, the gas diffusivity of an Andosol with high porosity is higher than in a Gray Lowland soils, however the gas movement through macropores and cracks were not active in an Andosols (Osozawa 1998). On the one hand, the gas movement through macropores and cracks is dominant in a Gray Lowland soil (Osozawa 1998). Although the gas diffusivity estimated by the value of D/D_0 in the surface soil (0–0.1m) disturbed by the plowing of the Andosol was higher

than that of the Gray Lowland soil (Fig. 6), there were no differences in the value of D/D_0 at a depth of more than 0.3 m between the Andosol and the Gray Lowland soil. However, the value of D/D_0, which measured by 100 mL core, could not take into account the gas flowing through macropores and cracks. It suggests that the gas flowing in the subsoil of the Gray Lowland soil with macropores and cracks was higher than the value of D/D_0. It is considered that the gas diffusivity around macropores and cracks in the subsoil of the Gray Lowland soil might be higher than in the Andosol. Therefore, mobility of N_2O in the subsoil around the macropores and cracks of the Gray Lowland soil might be higher than that in the Andosol. Additionally, the N_2O produced in the subsoil of the Andosol without macropores and cracks might have been reduced to N_2 before it was emitted to the atmosphere. These results indicate that the difference in water mobility, NO_3^-, O_2, and N_2O in the soils, especially the subsoil, between the Gray Lowland and the Andosol might be the reason for variation in the seasonal pattern of N_2O fluxes, the N_2O concentration profile in the soil, and the ratio of contribution of the subsoil to N_2O production in the soil between both soils (Figs. 7– 9, Table 2).

The CO_2 emitted from the soil into the atmosphere is produced by the respiration of plant roots and soil microbes (Smith et al. 2003). In spite of the differences in soil type, CO_2 could be produced in the top soil, because plant roots and soil microbes were distributed in the top soil (Nakamoto 1993, Osozawa 1998). More than 90 % of the CO_2 emitted from the soil to the atmosphere was produced in the top soil in both the soil types (Table3). Unlike N_2O, there was no significant correlation between the CO_2 flux through a 0.3 m depth and that from the soil into the atmosphere in both soils. In this way, our results corresponded to the previous reports (Nakamoto 1993, Osozawa 1998, Smith et al. 2003).

Conclusions

In the Gray Lowland soil and the Andosol, N_2O and CO_2 were mainly produced in the top soil (0–0.3m depth). The seasonal patterns of the CO_2 concentration profile were similar in both the soil types. However, the N_2O concentration profile in the soil was different between the two soils. Additionally, the ratio of contribution of the subsoil to the N_2O production in the soil of the Gray Lowland soil was higher than that of the Andosol, because the N_2O production in the subsoil around macropores and cracks of the Gray Lowland soil might have been activated by the leaching of NO_3^- through macropores and cracks. Subsequently the N_2O produced in the subsoil could have been rapidly emitted from the soil into the atmosphere through macropores and cracks. This suggests that the variations in the N_2O concentration profile between the two soils are caused by the differences in soil structure. This is especially because of the presence of macropores and cracks in the soil structure, which influenced the production and movement of N_2O in the soil.

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Table 1 Soil texture, structure, and saturated hydraulic conductivity of the study site

CL, ; HC, ; LiC, ; ND, not detected; SiC, ; SL, .

Horizon	Depth	Texture	Structure			Saturated hydraulic conductivity
	(m)		Grade	Size	Type	(m s^{-1})
Gray Lowland soil						
Ap	0-0.28	SiC	Strong	Medium	Subangular blockly	$1.0*10^{-7}$
B	0.28-0.48	SiC	Strong	Medium	Subangular blockly	$1.8*10^{-6}$
C1	0.48-0.68	HC	Strong	Coarse	Prismlike	$4.6*10^{-6}$
C2	0.68-1.0+	SiC	-	-	Massive	$2.2*10^{-4}$
Andosol						
Ap	0-0.3	CL	-	-	Granular	$3.3*10^{-6}$
AB	0.3-0.37	LiC	Moderate	Medium	Subangular blockly	$2.2*10^{-5}$
B	0.37-0.47	LiC	Moderate	Medium	Subangular blockly	$2.3*10^{-5}$
BC1	0.47-0.75	CL	Weak	Coarse	Subangular blockly	$4.3*10^{-5}$
BC2	0.75-0.9	LiC	Weak	Coarse	Subangular blockly	ND
C	0.9-1.0+	SL	-	-	Massive	ND

Table 2 Cumulative flux, production, and mass balance of N_2O during the study period

Year	Period	Cumulative N_2O flux during study period		Mass of N_2O in the topsoil (above 0.3m)		N_2O production by topsoil	Contribution ratio of topsoil
		Surface flux (F_0)	Through 0.3 m ($F_{0.3}$)	Beginning (Ms)	End (Me)	(P)	(P/F_0)
		(mg N m^{-2})		(mg N m^{-2})		(mg N m^{-2})	
	Gray Lowland soil						
1995	6/13-10/28	760 ± 140	86 ± 6	0.35	0.55	670	0.89
1996	7/2-10/31	310 ± 33	71 ± 4	0.26	1.02	240	0.77
1997	6/13-10/23	450 ± 200	56 ± 5	0.32	0.56	390	0.88
1998	6/23-10/27	430 ± 76	65 ± 4	0.20	0.83	370	0.85
1999	5/26-10/20	930 ± 250	80 ± 7	0.69	0.64	850	0.91
2000	5/30-10/24	1190 ± 450	160 ± 10	0.21	0.70	1030	0.86
	Average	680 ± 96	87 ± 3	0.34	0.72	590	0.86
	Ansosol						
1998	6/15-9/29	630 ± 88	1.3 ± 0.1	0.23	8.37	640	1.01
1999	6/6-9/13	1980 ± 230	6.2 ± 0.6	0.12	0.30	1980	1.00
2000	7/17-9/18	1430 ± 140	7.8 ± 1.1	0.20	0.33	1420	0.99
	Average	1350 ± 94	5.1 ± 0.4	0.18	3.00	1350	1.00

F_0 was measured using the chamber method and $F_{0.3}$ was measured using the gradient method. Values are mean ± standard deviation.

Table 3 Cumulative flux, production, and mass balance of CO_2 during the study period

Year	Period	Cumulative CO_2 flux during the study period		Mass of CO_2 in the topsoil (above 0.3m)		CO_2 production by topsoil	Contribution ratio of topsoil
		Surface flux (F_0)	Through 0.3 m ($F_{0.3}$)	Beginning (Ms)	End (Me)	(P)	(P/F_0)
		($g\ C\ m^{-2}$)		($g\ C\ m^{-2}$)		($g\ C\ m^{-2}$)	
	Gray Lowland soil						
1998	6/23-10/27	360 ± 29	22 ± 1	0.32	0.23	330	0.94
1999	5/26-10/20	410 ± 26	44 ± 2	0.12	0.13	370	0.89
2000	5/30-10/24	430 ± 27	16 ± 1	0.12	0.16	410	0.96
	Average	400 ± 16	27 ± 1	0.22	0.22	370	0.93
	Andosol						
1998	6/15-9/29	380 ± 9	4.8 ± 0.1	0.34	0.70	380	0.99
1999	6/6-9/13	540 ± 9	7.1 ± 0.3	0.25	1.31	530	0.99
2000	7/17-9/18	340 ± 6	3.7 ± 0.3	0.53	0.47	320	0.99
	Average	420 ± 5	5.2 ± 0.1	0.37	0.83	410	0.99

F_0 was measured using the chamber method and $F_{0.3}$ was measured using the gradient method. Values are mean ± standard deviation.

Table4 The regression curves of the soil water suction (pF)–D/D_0 used to the calculation of the gas fluxes in the soil profile.

Depth (m)	Regression curve	R^2	Mean square of residual
Gray Lowland soil			
0.15–0.20	$Y= 4.6\times10^{-4}x^3-2.2\times10^{-3}x^2+3.5\times10^{-3}x+6.0\times10^{-5}$	0.973	5.0×10^{-8}
0.23–0.28	$Y= 1.0\times10^{-3}x^3-3.7\times10^{-3}x^2+4.9\times10^{-3}x-2.0\times10^{-5}$	0.972	2.0×10^{-7}
0.32–0.37	$Y= 9.8\times10^{-4}x^3-3.5\times10^{-3}x^2+4.7\times10^{-3}x-2.0\times10^{-5}$	0.999	7.8×10^{-9}
0.43–0.48	$Y= 1.1\times10^{-3}x^3-3.8\times10^{-3}x^2+5.3\times10^{-3}x-1.0\times10^{-5}$	0.991	1.4×10^{-7}
0.54–0.59	$Y= 4.0\times10^{-3}x^3-1.1\times10^{-2}x^2+9.5\times10^{-3}x-1.7\times10^{-4}$	0.994	4.3×10^{-7}
Andosol			
0.13–0.18	$Y= -8.0\times10^{-5}x^3+2.2\times10^{-3}x^2-3.4\times10^{-3}x+1.3\times10^{-3}$	0.998	1.0×10^{-7}
0.31–0.36	$Y= -5.2\times10^{-4}x^3+4.2\times10^{-3}x^2-6.7\times10^{-4}x-1.2\times10^{-3}$	0.993	1.3×10^{-6}
0.40–0.45	$Y= -9.6\times10^{-4}x^3+5.8\times10^{-3}x^2-1.6\times10^{-3}x-1.1\times10^{-3}$	0.983	2.9×10^{-6}
0.58–0.63	$Y= -5.0\times10^{-5}x^3+8.6\times10^{-4}x^2+1.8\times10^{-3}x-1.2\times10^{-3}$	0.993	3.5×10^{-7}

Y denotes the value of D/D_0, x denote the soil water suction (pF), pF = log (-10.2 φ), φ is the soil water suction (kPa)

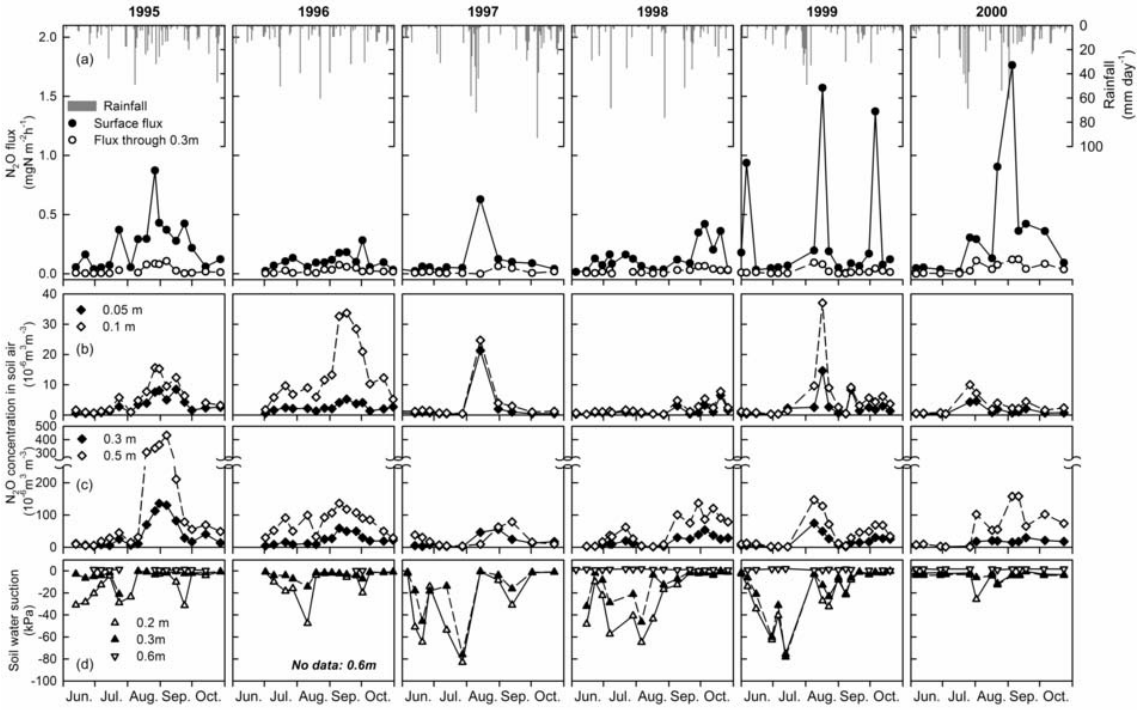

Figure 1 The seasonal patterns of (a) rainfall and N_2O fluxes from the soil surface and through a 0.3 m depth in the soil profile, (b, c) N_2O concentration in soil air (at 0.05, 0.1, 0.3, and 0.5 m depths), (d) soil water suction (at 0.2, 0.3, and 0.6 m depths) in the Gray Lowland soil from 1995 to 2000. Chemical fertilizer was applied at the end of April. The surface fluxes were reported by Kusa *et al.* 2002.

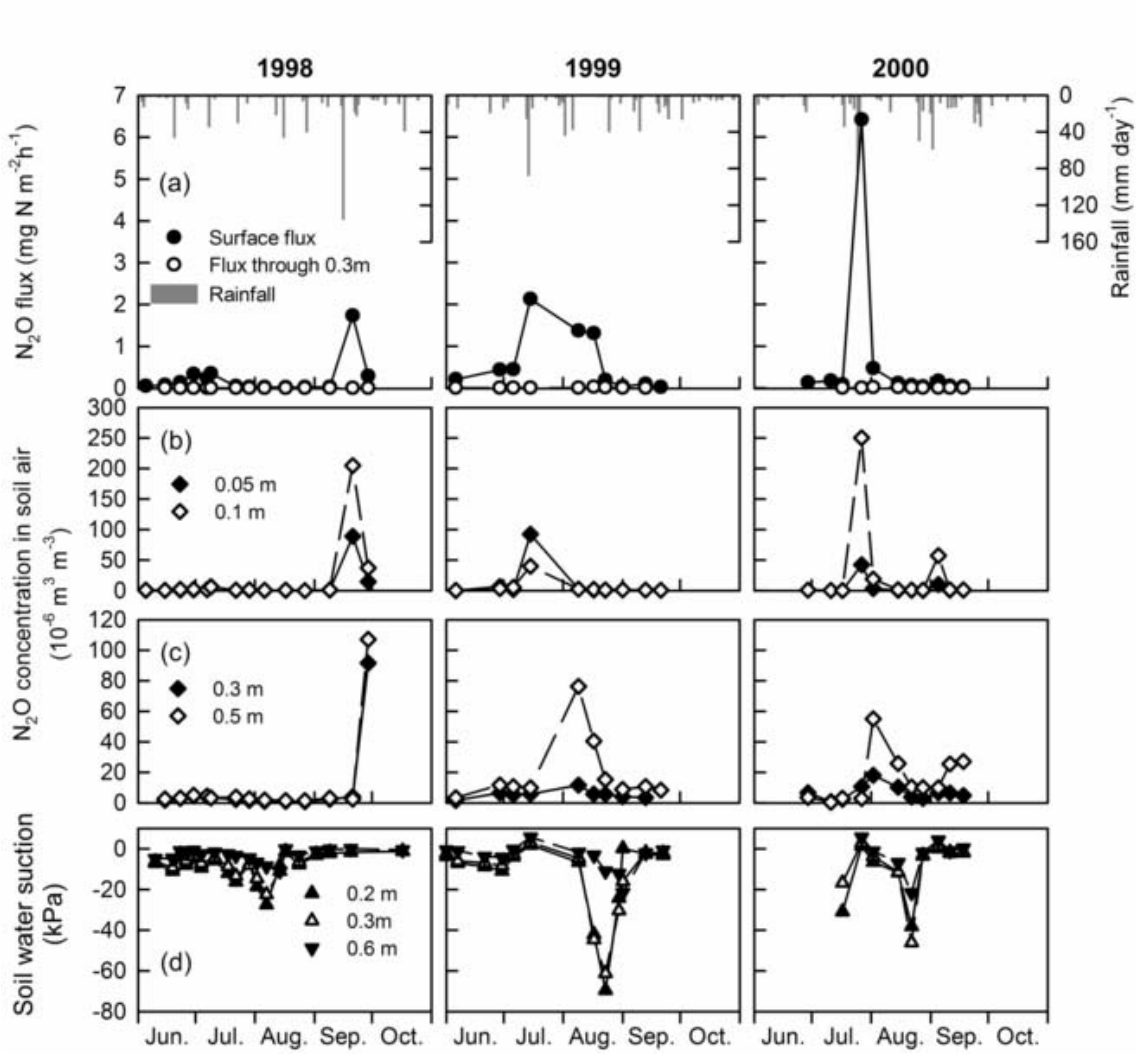

Figure 2 The seasonal pattern of (a) rainfall and N_2O fluxes from the soil surface and through a 0.3 m depth in the soil profile, (b, c) N_2O concentration in soil air (at 0.05, 0.1, 0.3 and 0.5 m depths), (d) soil water suction (at 0.2, 0.3 and 0.6 m depths) in the Andosol from 1998 to 2000. Chemical fertilizer was applied in mid-May. The surface fluxes were reported by Kusa *et al.* 2006.

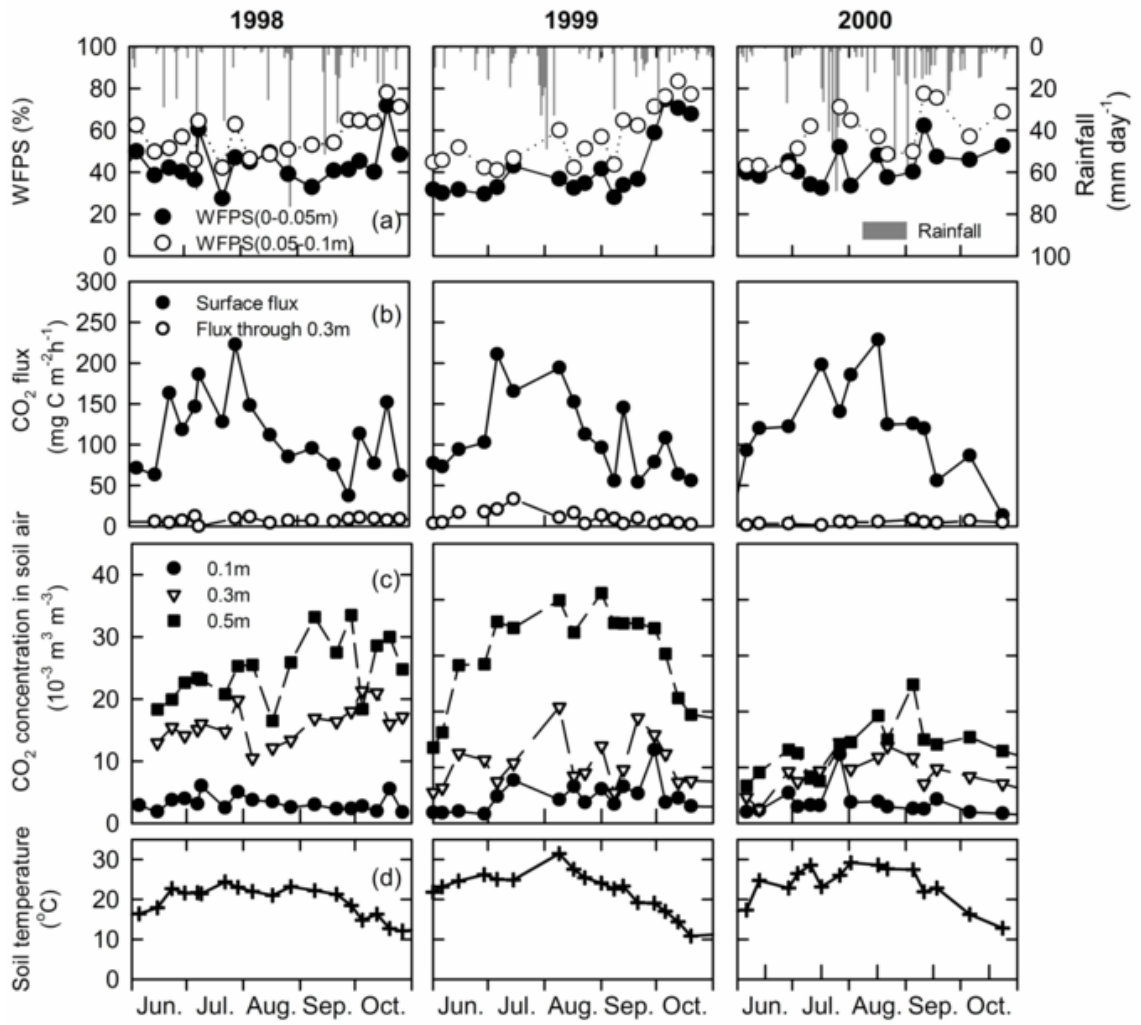

Figure 3 The seasonal pattern of (a) rainfall and WFPS of the soil surface, (b) CO_2 fluxes from the soil surface and through a 0.3 m depth in the soil profile, (c) CO_2 concentrations of soil air (0.1, 0.3 and 0.5 m depths), (d) soil temperature at a 0.1 m depth from the Gray Lowland soil from 1998 to 2000. WFPS (0-0.05 m) and the soil temperature were reported by Kusa *et al.* 2002.

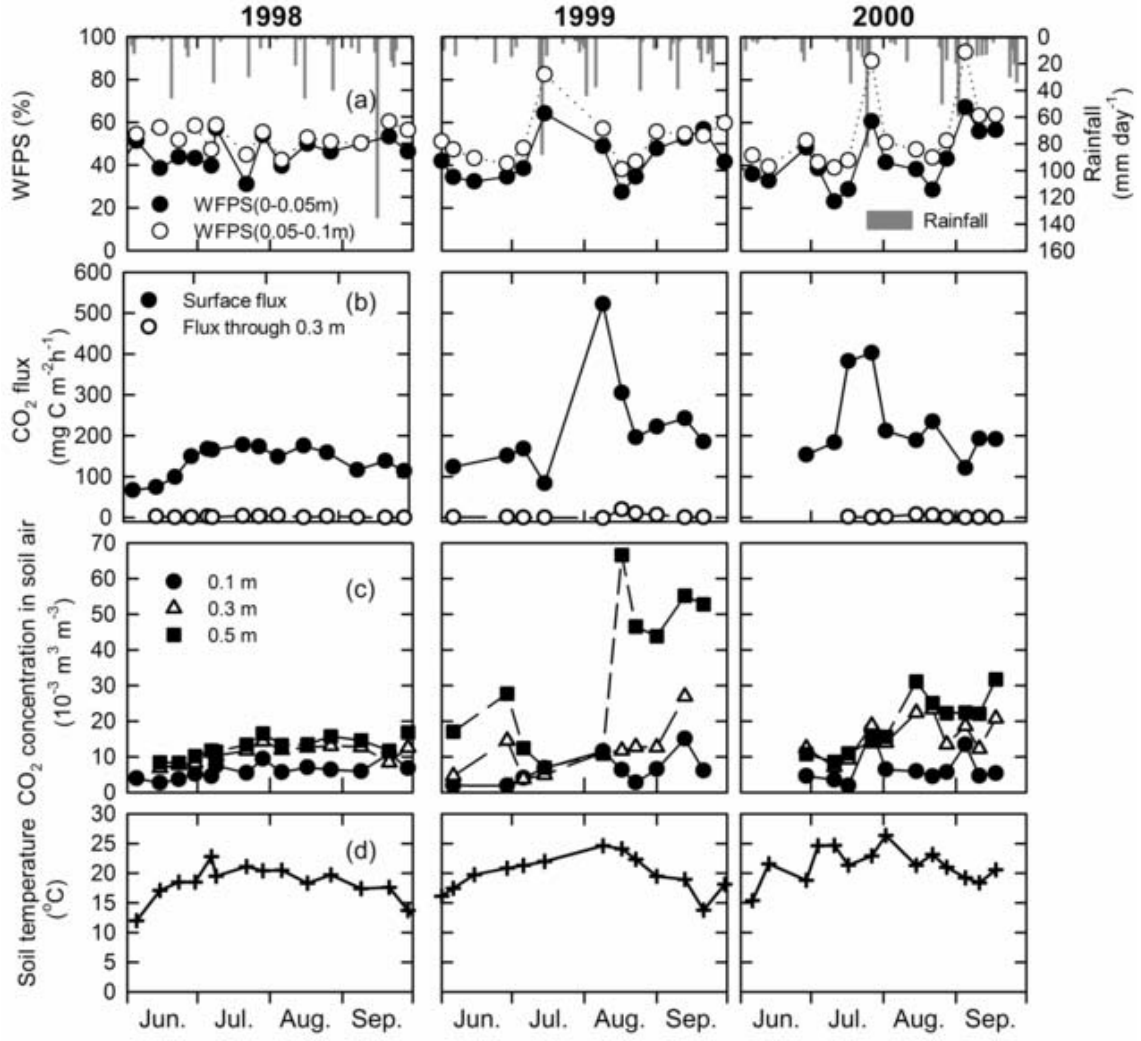

Figure 4 The seasonal pattern of (a) rainfall and WFPS in the soil surface, (b) CO_2 fluxes from the soil surface and through a 0.3 m depth in the soil profile, (c) CO_2 concentrations in soil air (at 0.1, 0.3 and 0.5 m depths), (d) soil temperature at a 0.01 m depth from the Andosol from 1998 to 2000. WFPS (0-0.05m), the surface fluxes and the soil temperature were reported by Kusa *et al.* 2006.

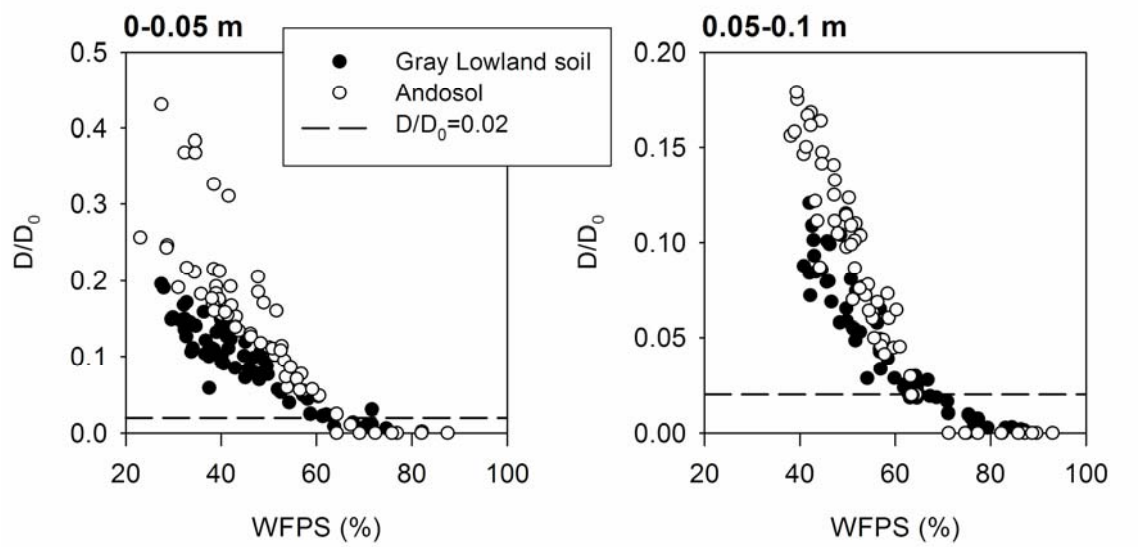

Figure 5 The relationships between the relative gas coefficient (D/D_0) and soil moisture (WFPS) from 1998 to 2000. WFPS (0-0.05m) was reported by Kusa *et al.* 2002 and 2006.

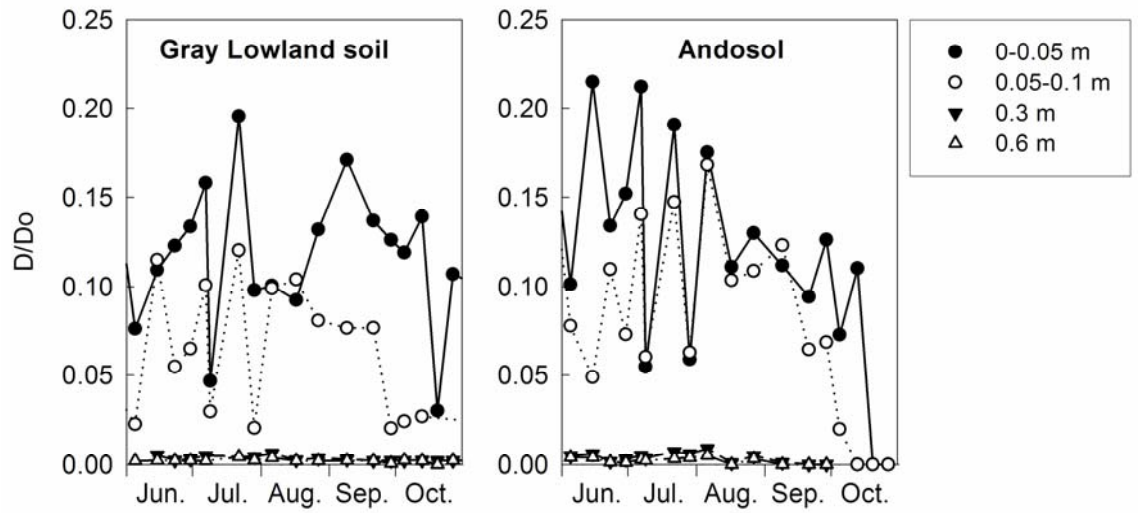

Figure 6 Seasonal patterns of the relative gas coefficient (D/D_0) for the Gray Lowland soil and the Andosol in 1998.

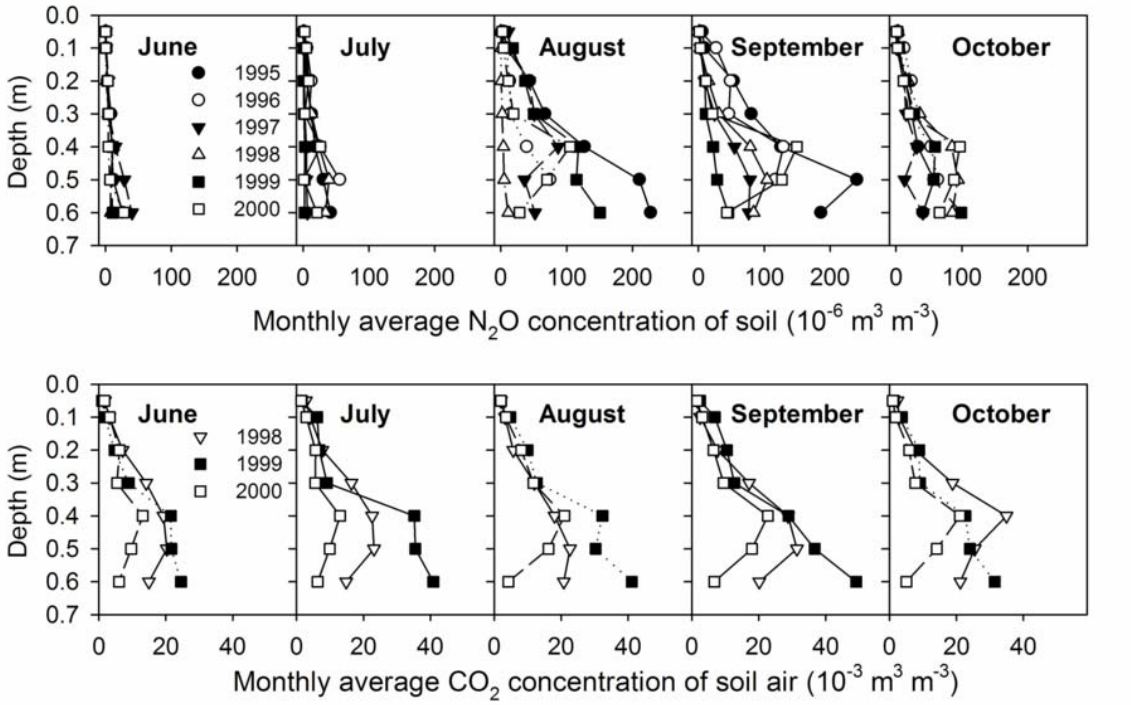

Figure 7 Monthly average concentrations of N_2O and CO_2 in the soil profile from the Gray Lowland soil from 1995 to 2000 (N_2O) and from 1998 to 2000 (CO_2).

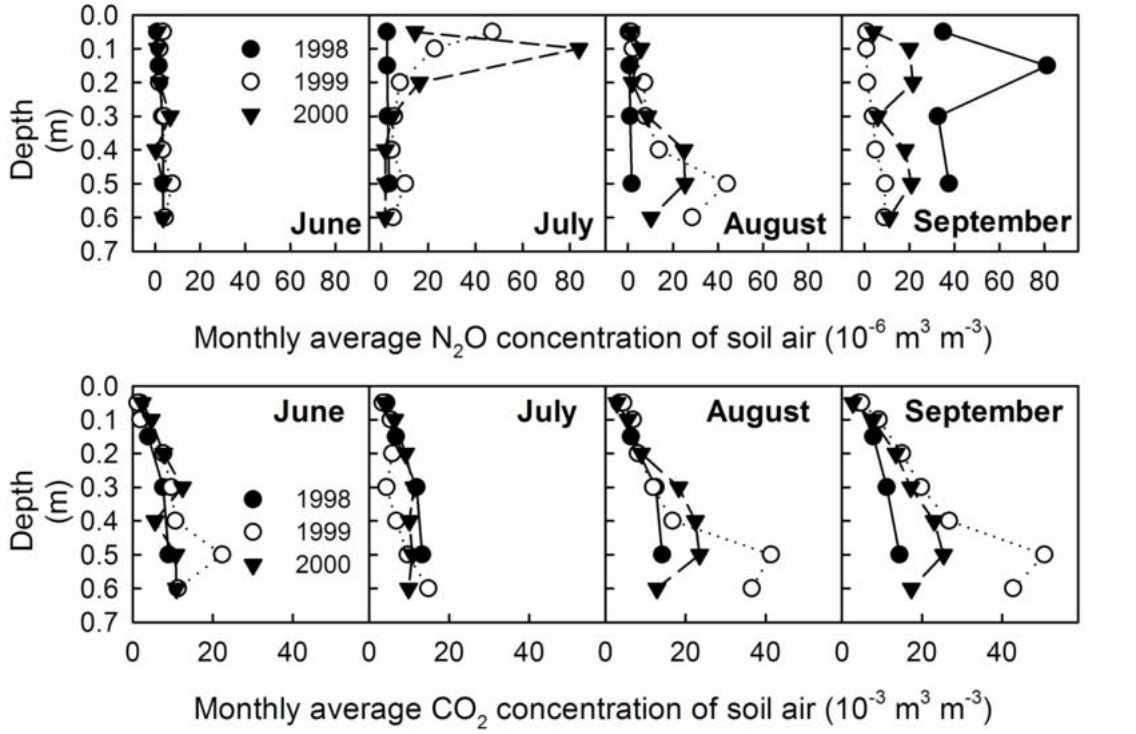

Figure 8 Monthly average concentrations of N_2O and CO_2 in the soil profile from the Andosol from 1998 to 2000.