Division: Environment

Contribution: Full-length paper

Title: Effects of environmental factors on temporal variation in annual carbon dioxide and nitrous oxide emissions from an unfertilized bare field on Gray Lowland soil in Mikasa, Hokkaido, Japan

Running head: Variation in CO$_2$ and N$_2$O emission from bare soil

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Abstract

Soil is one of the important sources of atmospheric carbon dioxide (CO$_2$) and nitrous oxide (N$_2$O). Study of CO$_2$ and N$_2$O emission from bare soil may explain the annual change of carbon (C) in soil organic matter (SOM) and help analyzing the N$_2$O production from SOM. Therefore, CO$_2$ and N$_2$O emissions associated with the decomposition of SOM from bare soil are important factors for assessing the C budget and N$_2$O emission in agricultural field. We conducted a study over seven years to assess the controlling factors of CO$_2$ and N$_2$O emissions from unplanted and unfertilized soil in Mikasa, Hokkaido, Japan.

Carbon dioxide flux increased in the summer, and there were significant positive correlations between the CO$_2$ flux and soil temperature in the first four years. However, apparent relationships between CO$_2$ flux and WFPS, soil NH$_4$ and NO$_3$ concentrations were not observed. The slope of monthly CO$_2$ emission against mean monthly temperature was positively correlated with monthly precipitation. These results suggest that response of CO$_2$ production by increase in soil temperature becomes more sensitive in wet soils. The average CO$_2$ emission during the study period was 2.53 Mg C ha$^{-1}$ yr$^{-1}$, and uncertainty of the annual CO$_2$ emission was 24%. Annual precipitation explained the yearly
Nitrous oxide flux increased from July to October, and was positively correlated with CO$_2$ flux. Based on the ratio of N$_2$O-N:NO-N of fluxes, N$_2$O appeared to be the main product of denitrification. The average N$_2$O emission in the study period was 4.88 kg N ha$^{-1}$ yr$^{-1}$, and uncertainty of annual N$_2$O emission was 58.5%. Strong relationships between the monthly emissions of CO$_2$ and N$_2$O suggest that N$_2$O production by denitrification is strongly affected by SOM decomposition. Unlike the CO$_2$ emission, the relationship between N$_2$O emission and precipitation was not observed because of the multiple pathways of nitrification and denitrification for N$_2$O production induced by SOM decomposition.

Key words: carbon dioxide, denitrification, nitrous oxide, soil organic matter decomposition, temporal variation.
INTRODUCTION

The contributions of atmospheric carbon dioxide (CO$_2$) and nitrous oxide (N$_2$O) to global warming are reported to be 60% and 6%, respectively. Since the Industrial Revolution, concentrations of these greenhouse gases in the atmosphere have increased at a rate of 1.4 ppm yr$^{-1}$ and 0.8 ppb yr$^{-1}$, respectively (Intergovernmental Panel on Climate Change (IPCC) 2007). The emission of carbon (C) that has originated due to the change in land use from 1980 to 1989 contributes 24% of the global annual CO$_2$ emission. Similarly, the emission of N$_2$O originating from agricultural fields in 1989 contributed 24% of the total global N$_2$O emission (Mosier et al. 1998). Recently, the annual N$_2$O emissions from fertilized cropland and grassland at a global level were estimated at 3.3 and 0.8 Tg N yr$^{-1}$, respectively (Stehfest & Bouwman 2006).

Some studies have reported that there is a reduction in C from agricultural soils because micro-organisms decompose soil organic matter (SOM) and emit CO$_2$ (Hu et al. 2004; Koga et al. 2006; Koizumi et al. 1993; Mu et al. 2006, 2008; Shimizu et al. 2009). The reduction of C from soil has been estimated by a difference in soil C over 20 years in a 0-30 cm soil surface (IPCC 2006; Paustian et al. 1997). In recent years, the global warming potential (GWP) has been used...
to evaluate the effect of agricultural activities on global warming (Chu et al. 2007; Jones et al. 2006; Koga et al. 2006; Mosier et al. 2005, Mu et al. 2006; Robertson & Grace 2004; Six et al. 2004). Therefore, it is necessary to estimate the annual reduction of soil C in order to evaluate the annual effect of agricultural activities on global warming. However, annual reduction of soil C is difficult to be detected from the investigation of annual change in the amount of soil C, because soil C is usually very large compared to C emission from the soil. In agricultural fields, decomposition of SOM can be measured as the CO$_2$ emission from bare fields in which root respiration is excluded (Hanson et al. 2000; Hu et al. 2004; Mu et al. 2006, 2008; Shimizu et al. 2009; Subke et al. 2006). Since the decomposition of SOM is resulted from microbial activities, it is influenced by chemical and physical conditions such as soil temperature, water conditions, pH, and so on. Due to the spatial variability of those factors (Yanai et al. 2003), it is difficult to investigate the factors affecting the decomposition of SOM at the field level. Understanding the factors affecting the decomposition of SOM, quantifying the annual CO$_2$ emission from bare fields, and clarifying factors affecting the temporal variations in annual CO$_2$ emissions from an unfertilized bare soil are important to estimate the loss of C from soil. Those understandings will provide
beneficial information to the study of the effect of agricultural activities on C cycle or global warming at a long-term span such as eco-balance or life cycle analysis (Koga et al. 2006; Kimura et al. 2007).

Nitrous oxide is produced by nitrification and denitrification processes in the soil. Therefore, N\textsubscript{2}O emission is affected by the soil's chemical and physical conditions (Bremner 1997; Colbourn & Dowdell 1984; Mosier 1998; Stehfest & Bouwman 2006). Because ammonium (NH\textsubscript{4}\textsuperscript{+}) is used in the nitrification process and nitrate (NO\textsubscript{3}\textsuperscript{-}) in the denitrification process, the N\textsubscript{2}O emission from agricultural fields is usually in proportion to the N application rate (Bouwman 1996). Based on this, methods of estimating N\textsubscript{2}O emissions from agricultural fields have been proposed (IPCC 2006). In Tier 1 and 2 in the IPCC Guidelines for National Greenhouse Gas Inventories (IPCC 2006), the emission factor (EF) is the ratio of N-induced N\textsubscript{2}O emission, which is a difference between N\textsubscript{2}O emissions from N-fertilized and unfertilized field, divided by the amount of applied N fertilizer. However, to calculate the original EF values, the N\textsubscript{2}O emission originating from soil organic N should be considered, because N\textsubscript{2}O emission from the soil in agricultural fields is usually composed not only of N\textsubscript{2}O emissions originating from the applied N, but also of N\textsubscript{2}O emissions induced by
decomposition of SOM. The N\textsubscript{2}O emission from bare fields is regarded as the emission originating from the soil organic N. There are several studies that have reported annual N\textsubscript{2}O emissions from unfertilized bare fields (Akiyama et al. 2006; Clayton et al. 1997; van Groenigen et al. 2004; Kamp et al. 1998; Koga et al. 2004; Zou et al. 2005). In addition, a temporal variation in N\textsubscript{2}O emission from agricultural fields has been reported by several studies (Drury et al. 2006; Kusa et al. 2002; Takakai et al. 2006; Zou et al. 2005). In any case, however, the reported studies have not focused on the susceptible factors for N\textsubscript{2}O flux or annual N\textsubscript{2}O emission from SOM. There are few studies focusing on analysis of mechanisms of N\textsubscript{2}O production and emission induced by SOM decomposition in agricultural bare fields at a long-term span. The knowledge about the controlling factors of N\textsubscript{2}O flux and annual emissions will be also useful for the study of the effect of agricultural activities on C cycle or global warming in a long run.

The objective of this study was to clarify the controlling factors for CO\textsubscript{2} and N\textsubscript{2}O emissions from agricultural bare soil based on seven years of monitoring in central Hokkaido, Japan.

MATERIALS AND METHODS
Site description

This study was conducted in Mikasa, central Hokkaido, Japan (43° 14.4′ N, 141° 50′ E). The soil type is Gray Lowland soil (Gleysol; FAO/UNESCO). The mean annual temperature of the study site is 7.5°C and the mean annual precipitation is 1164 mm, of which 32% is in the form of snow. The soil does not freeze during the winter. Temperature usually increases in August and precipitation increases from July to October (Fig. 1). At a depth of 0–10 cm, the soil pH (H₂O) was 5.8 and the cation exchange capacity was 25.5 cmolc kg⁻¹. The soil was comprised of sand (12.1%), silt (51.2%) and clay (36.7%). Concentrations of soil C and N were 32.1 and 2.8 g kg⁻¹, respectively (Toma & Hatano 2007) and the C:N ratio was 11.5 at a depth of 0–10 cm in 1996. Soil C density was recorded as 108 Mg C ha⁻¹ at a 0-30 cm depth.

Experimental design

We set up an unfertilized bare plot (5 m × 8 m) inside a 2-ha onion field owned by a farmer in Mikasa in 1999 and conducted monitoring in 2000, from 2002 to 2005, and in 2007. In 2006, the unfertilized bare plot was shifted to another location in the same field by the farmer due to unavoidable circumstances.
Conventional management practices for onion cultivation were carried out on the studied plot every year, although chemical and organic fertilizers or residues were not applied. Most of weed was removed from the monitoring plot regularly. Onion is usually cultivated from May to September in Mikasa. The soil is usually tilled before fertilization and plowed up to a depth of 15 cm at the end of April. Onion roots are cut for harvest from mid to late August. The onion harvest is followed by plowing, which is carried out to incorporate the residues into the soil from the end of September to early October.

**Measurement of CO₂, N₂O, and NO fluxes**

Fluxes of CO₂, N₂O, and NO were measured by a closed chamber method (three or four replications) using two types of cylindrical stainless steel chambers from 10:00 am to 2:00 pm. In 2000, chambers of 30 cm in diameter and 35 cm in height were used (Kusa et al. 2002, 2006), whereas chambers of 20 cm in diameter and 25 cm in height were used from 2002 to 2007. The cover of the chamber was made of acryl and was equipped with a sample collector, a pressure regulating bag and a Tedlar bag (0.5 L). In 2000 and from 2002 to 2004, we inserted the chambers directly into the soil to a 2 cm depth and started
making measurements after 15 min. However, in 2005, 2006, and 2007, we used a chamber-base made of stainless steel with a diameter of 20 cm. The upper part of the chamber-base had a slight depression, which was filled with water to seal it during the measurement (Toma & Hatano 2007; Toma et al. 2007). The base was kept on the ground, except during plowing and was reset one day before the next sampling.

Gas samples were taken at time 0 min from inside the chamber and 6 min. (for CO$_2$) or 15 min. (for N$_2$O and NO) after closing the chamber (Nakano et al. 2004; Toma & Hatano 2007). Using a 25 mL syringe, gas sample was taken ten times (total volume of 250 mL), and the gas sample was injected into a Tedlar bag (0.5 L). The bags were then brought to the laboratory and 20 mL of each gas sample was immediately transferred into glass vials (10 mL). From the samples of the bags, CO$_2$ and NO were analyzed using an Infrared CO$_2$ Analyzer (Model ZFP5YA3I, Fuji Electric, Tokyo, Japan) and a Chemoluminescence Nitrogen Oxide Analyzer (Model 265P, Kimoto Electric, Osaka, Japan), respectively. From the samples of the vials, N$_2$O was analyzed with an ECD Gas Chromatograph (Model GC-14B, Shimadzu, Kyoto, Japan).

The gas fluxes were calculated using the following equation:
\[ F = \rho \times \frac{V}{A} \times \frac{\Delta c}{\Delta t} \times \left[ \frac{273}{(273 + T)} \right] \times \left( \frac{P}{760} \right) \]

where, \( F \) is the flux (mg N or C m\(^{-2}\) h\(^{-1}\)), \( \rho \) is the gas density (\( \rho \) N\(_2\)O-N = 1.259 \( \times 10^6 \) and \( \rho \) CO\(_2\)-C = 0.536 \( \times 10^6 \) and \( \rho \) NO-N = 0.625 \( \times 10^6 \) mg m\(^{-3}\)), \( V \) is the volume of the chamber (m\(^3\)), \( A \) is the area of the chamber (m\(^2\)), \( \Delta c/\Delta t \) is the ratio of change in the gas concentration inside the chamber (10\(^{-6}\) m\(^3\) m\(^{-3}\) h\(^{-1}\)), \( T \) is the air temperature inside the chamber (\( ^\circ \)C), and \( P \) is the air pressure (mm Hg; see Kusa et al. 2002; Toma & Hatano 2007).

A positive value of \( F \) indicates gas emission from the soil to the atmosphere, while the negative value indicates gas uptake by the soil from the atmosphere.

Considering the machinery precision, N\(_2\)O flux values within the range from -6 to 6 \( \mu \)g N m\(^{-2}\) h\(^{-1}\), and CO\(_2\) flux values from -3.7 to 3.7 mg C m\(^{-1}\) h\(^{-1}\) and NO flux values from -0.2 to 0.2 \( \mu \)g N m\(^{-2}\) h\(^{-1}\) were regarded as 0 \( \mu \)g N or C m\(^{-2}\) h\(^{-1}\). The annual CO\(_2\) and N\(_2\)O emissions were calculated assuming linear changes between two sampling occasions.

The value of \( Q_{10} \), which is a relative increase of CO\(_2\) flux for 10\(^\circ\)C change in soil temperature, was calculated based on the Arrhenius equation and following equation (Hu et al. 2004):

\[ k = A \times \exp\left(\frac{-E}{RT}\right) \rightarrow \ln k = \ln A - \frac{E}{RT} \]
\[ Q_{10} = \frac{A \times \exp(-E / RT)}{A \times \exp(-E / R(T + 10))} \]

where, \( k \) is the CO\(_2\) flux, \( A \) is a constant, \( E \) is the activation energy, \( R \) is the universal gas constant (8.3144 J mol\(^{-1}\) K), and \( T \) is the absolute temperature (K).

The logarithm of the CO\(_2\) flux and the reciprocal of absolute temperature of soil were plotted, and \( E \) was determined from the slope. The value of \( Q_{10} \) was calculated by using \( T = 283 \) (K), because mean monthly air temperatures in this study site ranged from 10°C to 20°C (Fig. 1c and d).

**Measurement of other variables and frequency of sampling**

The soil temperature at a 5 cm depth was measured five times at the same time of measuring CO\(_2\), N\(_2\)O, and NO fluxes. Disturbed soil samples (0-5 cm depth) were taken at five replications and were combined for one composite sample.

Soil solutions were extracted by using distilled water through the study period and NH\(_4^+\)-N, NO\(_3^-\)-N and water soluble organic carbon (WSOC) concentrations were analyzed by colorimetry with indophenol-blue, ion chromatography (QIC Analyzer, Dionex, Osaka, Japan) and total organic carbon analyzer (Model TOC-5000A, Shimadzu, Kyoto, Japan), respectively. In 2007, soil NH\(_4^+\)-N
concentration extracted by distilled water was not measured, and in 2006 and
2007, soil NH$_4^+$-N concentration extracted by 2M KCl were analyzed. The
WSOC was not measured in 2000. The undisturbed soil samples were collected
by using a steel cylinder (100 mL) at three replicates and the water-filled pore
space (WFPS) was measured. The daily or annual meteorological data were
obtained from the local Iwamizawa Weather Station (43°12.6′N, 141°47.3′E).

After plowing, all samples were collected three times a week from May to June,
three times a month from July to August and once or twice a month during the
snow-cover season. During the period from snowmelt to onion cultivation and
from mid-October to snow cover, sampling was conducted two or three times per
month.

Statistical analyses

Relationships between CO$_2$ and N$_2$O fluxes and soil physical and chemical
properties, or annual CO$_2$ and N$_2$O emissions and of mean annual air
temperature or annual precipitation were analyzed by the linear or quadratic
curve regression. Variation in annual CO$_2$ or N$_2$O emissions among the study
period was evaluated by Tukey-Kramer test. The least significant difference test
was used to determine significant differences (\( P <0.05 \)).

Uncertainties were calculated using the following equation:

\[
\text{Uncertainty (\%)} = \frac{(\text{two-sided 95\% confidence interval})}{2}/\text{means} \times 100
\]

The two-sided 95\% confidence interval of CO\(_2\) and N\(_2\)O emissions were calculated by using the following equation:

\[
\text{Two-sided 95\% confidence interval} = t(\text{d.f., 0.05}) \times \text{standard error} \times 2
\]

where, d.f. is the degree of freedom, and \( t(\text{d.f., 0.05}) \) is the \( t \) value at 5\% significance level with two-sided alternative.

RESULTS

Seasonal variation in CO\(_2\) and N\(_2\)O fluxes

The CO\(_2\) flux peaked from August to October (Fig. 2a, 3a), whereas the N\(_2\)O flux increased from the end of July to October in all years (Fig. 2b, 3b). Soil temperature increased in August and this trend was similar to that of air temperature (Fig. 4a,f). The WFPS was high in early April and November, and low in August (Fig. 4b,g). From August to October, the WFPS was as low as around 50\%. Soil NH\(_4^+\)-N concentrations extracted by distilled water ranged from
0 to 7.6 mg N kg\(^{-1}\) each year and did not exhibit any consistent seasonal change (Fig. 4c,h). Although NH\(_4\)^+-N concentration extracted by 2M KCl in 2006 and 2007 did not show a distinct seasonal change (Fig. 4i), the range of concentrations (1.67-11.3 mg N kg\(^{-1}\) in 2006, 0.84-29.9 mg N kg\(^{-1}\) in 2007) was higher than the values obtained through extraction by water. Soil NO\(_3\)^--N concentrations increased from August to September each year (Fig. 4d,j). Fluctuation in soil WSOC concentrations was small throughout the year (Fig. 4e,k).

Correlation coefficients between the CO\(_2\) or N\(_2\)O fluxes and soil physical and chemical properties are given in Table 1. The CO\(_2\) flux was significantly and positively correlated with soil temperature from 2000 to 2004 (Table 1), and reached its maximum at 48.8% of WFPS (Fig. 5a). Although the N\(_2\)O flux was significantly and positively correlated with soil temperature in 2000 and 2004, N\(_2\)O flux was significantly and positively correlated with CO\(_2\) flux from 2000 to 2006. Carbon dioxide and N\(_2\)O fluxes were significantly correlated with soil NO\(_3\)^--N concentration, and N\(_2\)O flux was significantly correlated with soil temperature in 2000 and 2004. However, no significant correlation was observed between CO\(_2\) or N\(_2\)O fluxes and soil chemical or physical properties throughout the study period. There were no significant correlations between N\(_2\)O flux and
WFPS during the study period except for 2003, but N\textsubscript{2}O showed high fluxes when WFPS ranged from 30 to 60 % and reached its maximum at 48.4% (Fig. 5b).

The relationship between N\textsubscript{2}O flux and N\textsubscript{2}O-N:NO-N ratio is given in Fig. 6. The N\textsubscript{2}O flux increased with an increase in N\textsubscript{2}O-N:NO-N ratio, with 18 out of 49 samples of the N\textsubscript{2}O flux over 0.079 mg N m\textsuperscript{-2} h\textsuperscript{-1}, which resulted in the ratio of N\textsubscript{2}O-N:NO-N over 100. The average N\textsubscript{2}O flux was 0.079 mg N m\textsuperscript{-2} h\textsuperscript{-1}.

Therefore, about 37% of the N\textsubscript{2}O fluxes were higher than the average value.

Cumulative CO\textsubscript{2} and N\textsubscript{2}O emissions

Table 2 shows the annual CO\textsubscript{2} and N\textsubscript{2}O emissions and Q\textsubscript{10} values. The annual CO\textsubscript{2} emission ranged from 2.04 to 3.32 Mg C ha\textsuperscript{-1} yr\textsuperscript{-1}, and were significantly correlated with annual precipitation (Fig. 7, y = 0.0021 x -0.0499, R = 0.98, P < 0.01). The average CO\textsubscript{2} emission during the study period was 2.53 Mg C ha\textsuperscript{-1} yr\textsuperscript{-1}, and uncertainty of annual CO\textsubscript{2} emission was 24 %. The Q\textsubscript{10} values ranged from 1.11 to 2.38. There was no significant correlation between Q\textsubscript{10} value and annual precipitation (y = 0.0012x + 0.1918, R = 0.56, P = 0.11). However, the slope of the monthly CO\textsubscript{2} emission against mean monthly temperature from April
to October was significantly correlated with precipitation from April to October (Fig. 8, y = 0.0688x - 27.67, R = 0.90, P < 0.01). Carbon dioxide flux in this study was measured only during the daytime, and annual CO₂ emission was calculated assuming linear changes between two sampling occasions. This indicated that the decrease in CO₂ flux during the night time, when temperature decreased, was not considered in calculating the annual CO₂ emission from 2000 to 2004. The difference in maximum and minimum air temperature from July to September, which was the period of high air temperature, was below 10°C (8.8°C). In addition, the average Q₁₀ value from 2000 to 2004 was 1.99 (Table 2). If we assume that the CO₂ flux during the maximum daily air temperature is 1.99 times higher than the CO₂ flux during minimum daily air temperature, the measured annual CO₂ emission would be 1.33 times higher than the annual CO₂ emission that was calculated by considering the daily change in the CO₂ flux.

The annual N₂O emission ranged from 1.62 to 12.1 kg N ha⁻¹ yr⁻¹. The average N₂O emission during the study period was 4.88 kg N ha⁻¹ yr⁻¹, and uncertainty of the annual N₂O emission was 58.5 %. Uncertainty of yearly variation of CO₂ and N₂O emissions were 17.9 and 76.1 %, respectively. Although correlations
between N₂O emission and annual air temperature or precipitation were not significant, N₂O emission increased with an increase in CO₂ emission (Fig. 9).

Significant differences in annual CO₂ and N₂O emissions among the study years indicate yearly variation of CO₂ and N₂O emission in the study site (Table 2).

**DISCUSSION**

**Key factors determining the CO₂ flux in bare soil**

Several studies reported that CO₂ flux from soil showed an exponential increase with an increase in soil temperature (Boone *et al*. 1998; Hu *et al*. 2001; Jones *et al*. 2006; Schindlbacher *et al*. 2009). In this study, there were significant positive correlations between CO₂ flux and soil temperature (in 2000, 2002-2004) (Table 1). On the other hand, there was no significant correlation between CO₂ flux and WFPS except in 2003 (Table 1). Gulledge and Schimel (1998) reported that microbial respiration was directly proportional to the water holding capacity (10-60%). Moreover, Linn and Doran (1984) and Gulledge and Schimel (1998) reported that the maximum CO₂ production by microorganisms was at 60% WFPS. In our study, CO₂ flux peaked at 49.1% of WFPS. Therefore, soil moisture might not have influence on the production or emission of CO₂ linearly,
and there might be an appropriate condition of soil moisture for CO$_2$ production relating to the ventilation of soil. On the other hand, significant positive correlations between annual CO$_2$ emission and annual precipitation and between the slope of monthly CO$_2$ emission against mean monthly temperature from April to October and precipitation from April to October suggested that the production of CO$_2$ was greater in wet conditions even if temperatures were similar (Fig. 7, 8). Therefore, CO$_2$ production increased with an increase in soil temperature, but CO$_2$ production at the same temperature was more enhanced in wet conditions. High moisture content prevents soil gas from diffusing from soil to the atmosphere. This means that even if CO$_2$ was produced in soil in appropriate condition for CO$_2$ production, CO$_2$ might not necessarily be diffused at that time. There could be possibilities of high CO$_2$ flux to be detected during the season of high precipitation. Difference in timing between CO$_2$ production and diffusion from soil to the atmosphere could possibly be explained by a lack of correlation between CO$_2$ flux and WFPS and by a significant positive correlation between annual CO$_2$ emission and precipitation (Table 1, Fig. 7).

**Key factors determining the N$_2$O flux in bare soil**
Because N$_2$O is mainly produced by the processes of nitrification and denitrification in soil (Tiedje 1994), N$_2$O flux is often affected by soil NH$_4^+$-N or NO$_3^-$-N concentration (Livesley et al. 2009; Thornton & Valente 1996; Toma et al. 2007). However, no apparent correlation between N$_2$O flux and soil NH$_4^+$-N or NO$_3^-$-N concentration was observed in our study. Instead, we observed significant positive correlations between N$_2$O and CO$_2$ fluxes in all years, except for 2007 (Table 1). Similar positive correlations between N$_2$O and CO$_2$ fluxes were observed when crop residues were mixed with surface soil (Huang et al. 2004; Toma & Hatano 2007). In this study, the substrates for the nitrification and denitrification originated from SOM because no fertilizer was applied in the experimental plot, suggesting that N$_2$O production was strongly related with SOM decomposition. Therefore, one of the factors controlling the production of N$_2$O could be the supply of C and inorganic N from SOM decomposition. Some studies reported that soil temperature is one of the factors for N$_2$O production (Kamp et al. 1998; Mori et al. 2005; Smith et al. 1998; Tokuda & Hayatsu 2004). Since the CO$_2$ flux increased with an increase in soil temperature in this study (Table 1), soil temperature might not affect only the activity of N$_2$O production but also the supply of substrates for N$_2$O production. Bouwman (1990) summarized...
the results presented by Anderson and Levine (1986) and Lipchults et al. (1981) and reported that a value of \( \text{N}_2\text{O-N:NO-N} \) ratio below 1.0 indicates that \( \text{N}_2\text{O} \) was produced in the soil mainly by a nitrification process, while a value above 100 indicates that denitrification was the dominant process for \( \text{N}_2\text{O} \) production. In our study, About 37% of the \( \text{N}_2\text{O} \) fluxes that were higher than the average value, therefore, occurred when there was a condition of \( \text{N}_2\text{O-N:NO-N} \) ratio to be above 100. Generally, an increase in soil moisture contributes to \( \text{N}_2\text{O} \) production by denitrification (Linn & Doran 1984). It was reported that a WFPS of 50-60% would be suitable for the decomposition of soil organic matter and that these conditions accelerated denitrification due to the consumption of oxygen (Maag & Vinther 1999). Moreover, WFPS of 50-60% was the range in which \( \text{N}_2\text{O} \) was mainly produced by the denitrification process (Davidson et al. 2000). In our study, WFPS increased from about 40 to 60% from the end of July to early October, and \( \text{N}_2\text{O} \) flux was highest when the WFPS was at 46.5% (Fig. 4, 5).

Sawamoto and Hatano (2000) reported that an increase in \( \text{N}_2\text{O} \) production in autumn might be caused by nitrate transportation and diffusion inside soil aggregates after rainfall and development of denitrification areas inside the aggregates of well-structured soil in the same field of our study site. Therefore,
there is a possibility of aerobic and anaerobic conditions occurring simultaneously inside the aggregates and inter-aggregate pores in the well-structured soil in this study. This would suggest that high N$_2$O fluxes were induced not only by denitrification but also by nitrification. Nitrous oxide was the main product of the denitrification process in this study. Nitrous oxide fluxes were significantly correlated with CO$_2$ flux, and production of CO$_2$ was affected by soil temperature and precipitation. However, a significant correlation between N$_2$O flux or annual N$_2$O emission and WFPS or annual precipitation could not be found. This might be due to a complex process of N$_2$O production in soil. In this study, the source of inorganic N might be mainly SOM. Therefore, NH$_4$ and NO$_3$, which are substrates for nitrification and denitrification, could have been produced by mineralization and nitrification processes in soil. In the study of N$_2$O emission from organic fertilizer, similar correlations between N$_2$O and CO$_2$ emission were observed (Hayakawa et al. 2009). When N originates from organic matter, decomposition of organic matter is probably essential for producing N$_2$O. In any case, significant correlation between CO$_2$ flux and N$_2$O flux or monthly emission of CO$_2$ and N$_2$O suggest that climate change, which will affect SOM decomposition, might greatly affect N$_2$O emission.
induced by SOM decomposition.

Annual CO$_2$ emission and its spatial and temporal variations

The annual CO$_2$ emission, associated with decomposition of SOM, ranged from 2.04 to 3.32 Mg C ha$^{-1}$ yr$^{-1}$ (Table 2). The values of CO$_2$ emission from 2000 to 2004, however, might have been overestimated about 1.33 times because daily variation in CO$_2$ flux was not considered. Therefore, if we consider the overestimation of CO$_2$ emission, annual CO$_2$ emission ranged from 1.11 to 2.95 Mg C ha$^{-1}$ yr$^{-1}$. Mu et al. (2006) reported that CO$_2$ emitted from SOM in bare treatments in several types of land use (e.g. wheat, maize, onion, etc.) ranged from 3.01 to 5.68 Mg C ha$^{-1}$ yr$^{-1}$ in Hokkaido, Japan. Jacinthe et al. (2002) reported that 4.37 Mg C ha$^{-1}$ yr$^{-1}$ of CO$_2$ was emitted from a bare plot in Ohio, USA. Decomposition of SOM in the two studies might have been overestimated similar to our study because they used the trapezoidal rule to calculate annual CO$_2$ emission. Koizumi et al. (1994) reported that heterotrophic respiration ranged from 7.16 to 10.5 Mg C ha$^{-1}$ yr$^{-1}$ in three double-cropping agro-ecosystems (rice-barley, peanut-wheat, and dentcorn-italian ryegrass). They determined the SOM decomposition as the difference between total soil
respiration and plant respiration by calculating soil and root respirations using continuous data of soil temperature and the correlations between soil temperature and soil or root respirations. Koga et al. (2006) reported average SOM decomposition at a 20-year time scale since 1981 as 1.04 to 1.34 Mg C ha\(^{-1}\) yr\(^{-1}\) from the difference in total soil C within a depth of 20 cm in Hokkaido, Japan. The decomposition rate of SOM reported by Koizumi et al. (1994) and Koga et al. (2006) might not have been overestimated, and were larger and smaller than the values in this study, respectively. The reason of the difference in SOM decomposition may be due to the variation in location, cultivation system, and soil type of the study site. The mean annual temperature (13.1°C) of the study site reported by Koizumi et al. (1994) was higher than that in our study site. In addition, there were frequent tillage operations because of a double-cropping system. High temperature and frequency of plowing may have increased SOM decomposition in the study reported by Koizumi et al. (1994). On the other hand, the study site of Koga et al. (2006) was located in similar latitude (42°53′ N) as this study. Therefore, difference in SOM decomposition between our study and the study conducted by Koga et al. (2006) might be due to the differences in crop management tillage, and/or soil type. Mu et al. (2008) reported that CO\(_2\)
emission from bare soil reached to its maximum at 60-70% of the sum of silt and
clay content in soil. Moreover, Bellamy et al. (2005) reported that an amount of C
during 25-year was decreased with an increase soil C content. In these studies,
however, soil type of Andisols was not included. Further study about the
difference in SOM decomposition in various soil types will be required.

Carbon dioxide emission was measured at the same location in this study site
from 2000 to 2005. During this period, average annual CO$_2$ emission was 2.84
Mg C ha$^{-1}$ yr$^{-1}$, and 17 Mg C ha$^{-1}$ was estimated to be released from soil in 2000 -
2005. Because mass of C in surface (0-30cm) soil was 108 Mg C ha$^{-1}$, C loss of
17 Mg C ha$^{-1}$ corresponded to 15.7 % of surface soil C. We did not measure the
change in soil C storage during the study period. Therefore actual value of
reduction in soil C could not be shown in this study. Mass of soil C possibly could
be detected, if soil C change was measured during this study. Bellamy et al.
(2005), however, reported that loss of soil C was not detectable over 10 years
when soil C content was lower than 50 g kg$^{-1}$. Detecting the change in soil C in
this study site might be difficult. There are several ways of C input in agricultural
fields such as rainfall, weed, and alga. But most of weed or alga did not exist
during the study period. Carbon input from rainfall was not included in the IPCC
guideline for calculating soil C change (IPCC 2006). Thus, there might be no
major C inflow that supplemented soil C loss by SOM decomposition. Further
study would be required for matching the reduction of soil C and CO$_2$ emission
from unfertilized bare soil.

A large variation in CO$_2$ fluxes and annual CO$_2$ emissions among replication
were observed due to the spatial variation in CO$_2$ flux (Fig. 2a, 3a, and Table 2).
The average uncertainty of annual CO$_2$ emission (24 %) was higher than that of
annual CO$_2$ emission (17.9 %). The decomposition of SOM is generally affected
by soil properties (Koizumi et al. 1993; Mu et al. 2008), method of cultivation
(Koga et al. 2006), or land use practices (Mu et al. 2006). Mu et al. (2008)
reported a significant correlation between CO$_2$ emission and clay and silt content
in a bare field in the same district of our study site. In the study field, spatial
variation in total C content, soil C:N ratio, and microbial biomass C were reported
(Yanai et al. 2003). These factors possibly cause large uncertainty in annual CO$_2$
emission. Using a larger chamber or taking more replication for CO$_2$ flux
measurement might be required to improve an accuracy of annual CO$_2$
emissions.

Significant differences in annual CO$_2$ emissions from bare field during the
study period show that there is a yearly variation in annual CO\textsubscript{2} emission that is induced by the SOM decomposition (Table 2). A large uncertainty (17.9\%) of the average of annual CO\textsubscript{2} emission during the study period shows that continuous monitoring of CO\textsubscript{2} emission is required to determine the representative value of annual CO\textsubscript{2} emission. This yearly variation in annual CO\textsubscript{2} emission may suggest that climate will have a great impact on yearly CO\textsubscript{2} emission from SOM. Raich and Schlesinger (1992) reported that soil respiration, including root respiration, increased with an increase in annual precipitation or mean annual air temperature. Our study also showed that annual precipitation affect CO\textsubscript{2} emission from mineralization of SOM in the study site. Previous studies reported that the effect of chemical fertilizer application on microbial respiration is negligible (Ginting et al. 2003; Hu et al. 2004; Jacinthe et al. 2002). Hence, CO\textsubscript{2} emissions obtained in this study could be an approximation of SOM decomposition in a fertilized field. Several studies reported that soil C decreased in agricultural fields with low levels of organic matter application (Ginting et al. 2003; Hu et al. 2004; Jacinthe et al. 2002; Mu et al. 2008; Shimizu et al. 2009). The loss of soil C is reported to have a greater influence on global warming than the emissions of N\textsubscript{2}O or CH\textsubscript{4} (Jones et al.)
2006; Koga et al. 2006; Mosier et al. 2005; Mu et al. 2006). Although it is difficult to calculate an annual loss of C by measuring soil C reduction, it could possibly be done by measuring CO$_2$ emission from bare soil in an agricultural field. Assessing CO$_2$ emissions from bare fields, by calculating from the correlation between CO$_2$ emission and available environmental factors such as meteorological data or soil type, will be useful for cultivation management or planning of farm activities at a regional scale to mitigate C loss from soil.

Annual N$_2$O emission and its spatial and temporal variations

The annual N$_2$O emission in this study ranged from 1.62 to 12.1 kg N ha$^{-1}$ yr$^{-1}$ (average 4.88 kg N ha$^{-1}$ yr$^{-1}$). These values are higher than most reported values for bare fields (1.00 kg N ha$^{-1}$ yr$^{-1}$ (IPCC 2006), -0.02-0.13 kg N ha$^{-1}$ yr$^{-1}$ (Koga et al. 2004), 0.14-1.52 kg N ha$^{-1}$ yr$^{-1}$ (van Groenigen et al. 2004) or 0.36-0.14 kg N ha$^{-1}$ yr$^{-1}$ (Akiyama et al. 2006), 4.80 kg N ha$^{-1}$ yr$^{-1}$ (Kamp et al. 1998) or 4.25 kg N ha$^{-1}$ yr$^{-1}$ (Zou et al. 2005). The difference in annual N$_2$O emission might be due to soil physical and chemical properties. Nitrogen content in soil may affect N$_2$O production because there is no any N source for a substrate of N$_2$O in bare soil except for N in SOM. Klemedtsson et al. (2005) reported at significant negative
relationship between N\textsubscript{2}O emission and soil C:N ratio in forested peat soil in Europe, indicating that N\textsubscript{2}O emission increased with an increase in relative N to C in peat. Nitrogen contents of the soils in the study sites of Kamp \textit{et al.} (1998) (1.7 g kg\textsuperscript{-1}) and Zou \textit{et al.} (2005) (1.1 g kg\textsuperscript{-1}), in which N\textsubscript{2}O emissions exceeded 4.0 kg N ha\textsuperscript{-1}, were lower than the value of this study site (2.8 g kg\textsuperscript{-1}). Production of N\textsubscript{2}O in poorly drained soil increases due to an anaerobic condition in soil (van Groenigen \textit{et al.} 2004). Nitrous oxide emission reported by Koga \textit{et al.} (2004) was lower than the value of this study even if N content in their study site (2.4-3.1 g kg\textsuperscript{-1}) was similar to that of our study. This could possibly be due to the drainage quality of soil. Soil type in the study site of Koga \textit{et al.} (2004) is well-drained volcanic ash soil. Therefore, N concentration as a source of substrate for N\textsubscript{2}O and drainage condition that may control aerobic- and anaerobic condition of soil might be indicator of the potential N\textsubscript{2}O source from unfertilized bare field. Large variations in flux and annual emission of both N\textsubscript{2}O and CO\textsubscript{2} were observed in our study (Fig. 1b, 2b and Table 2). There is considerable uncertainty in the amount of N\textsubscript{2}O emission from unfertilized bare fields (Akiyama \textit{et al.} 2006; Zou \textit{et al.} 2005). Both CO\textsubscript{2} and N\textsubscript{2}O emissions were influenced by
soil properties (van Groenigen et al. 2004) and cultivation methods (Koga et al. 2004). In this study site, high spatial variability (CV 217%) of N$_2$O flux was reported (Yanai et al. 2003). Yanai et al. (2003) also reported that soil organic matter and soil pH were main soil-related determining factors for N$_2$O flux.

The temporal variation in N$_2$O was confirmed to be similar to that in some previous studies (Drury et al. 2006; Kusa et al. 2002; Takakai et al. 2006; Zou et al. 2005). In this study, neither N fertilizer nor crop residue was applied in the experimental plot from 1999 to 2005. Therefore, it is expected that the substrate required for production of N$_2$O had been reduced. However, the annual N$_2$O emission did not consistently decrease from 2000 to 2005, resulting in a large uncertainty of annual N$_2$O emission (Table 2). This suggests that the effect of climatic factors on N$_2$O emission might be greater than the reduction in substrate required for N$_2$O production. Annual N$_2$O emission tended to increase with an increase in annual precipitation. The relationship between annual N$_2$O emission and annual precipitation was not significant, whereas annual CO$_2$ emission was significantly correlated with annual precipitation (Fig. 7). The reason for this could be due to the fact that there is a difference in environmental factors that is required for mineralization, nitrification, and denitrification. In mineralization and
nitrification processes, oxygen is required for oxidation of organic C or NH$_4$.

However, an anaerobic condition is required for denitrification because NO$_3$ is used as an electron capture (Bouwman and Boumans 2001). Since N$_2$O was produced through these three biological processes in soil, a correlation between annual N$_2$O emission and annual precipitation may not be significant.
ACKNOWLEDGEMENTS

This study was financially supported by the Global Environmental Research Program of the Ministry of the Environment of Japan (No. S-2). The authors thank the farm owner Takeshi Morimoto for allowing us to conduct this study.
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Figure legends.

Figure 1 Seasonal variations in monthly precipitation [a, b] and mean monthly air temperature [c, d].

Figure 2 Seasonal variations in CO$_2$ [a], N$_2$O [b], and NO [c] fluxes in 2000, 2002, and 2003. Arrows indicate the timing of plowing.

Figure 3 Seasonal variations in CO$_2$ [a], N$_2$O [b], and NO [c] fluxes from 2004 to 2007. Arrows indicate the timing of plowing.

Figure 4 Seasonal variations in soil temperature [a & f], WFPS [b & g], soil NH$_4^+$-N [c, h & J], NO$_3^-$-N [d & i], and WSOC [e & j] concentrations.

Figure 5 Relationships between CO$_2$ or N$_2$O fluxes and WFPS.

Figure 6 Relationship between N$_2$O flux and N$_2$O-N:NO-N ratio.

Figure 7 Relationship between CO$_2$ emission and annual precipitation.
Figure 8 Relationship between slope of the monthly CO$_2$ emission against mean monthly temperature and precipitation from April to October.

Figure 9 Relationship between monthly CO$_2$ emission and N$_2$O emission from April to October.
Figure 1 Seasonal variations in monthly precipitation [a, b] and mean monthly air temperature [c, d].
Figure 2 Seasonal variations in CO\textsubscript{2} [a], N\textsubscript{2}O [b], and NO [c] fluxes in 2000, 2002, and 2003. Arrows indicate the timing of plowing.
Figure 3 Seasonal variations in CO$_2$ [a], N$_2$O [b], and NO [c] fluxes from 2004 to 2007. Arrows indicate the timing of plowing.
Figure 4  Seasonal variations in soil temperature [a & f], WFPS [b & g], soil NH$_4^+$-N [c, h & j], NO$_3^-$-N [d & i], and WSOC [e & j] concentrations.
Figure 5 Relationships between CO$_2$ or N$_2$O fluxes and WFPS.
Figure 6 Relationship between N$_2$O flux and N$_2$O-N:NO-N ratio.
Figure 7 Relationship between \( \text{CO}_2 \) emission and annual precipitation.

\[
y = 0.0021x - 0.0499 \\
R = 0.98, P < 0.01
\]
Figure 8 Relationship between slope of the monthly CO$_2$ emission against mean monthly temperature and precipitation from April to October.

\[ y = 0.0688x - 27.67 \]

\[ R = 0.90, \ P < 0.01 \]
Figure 9 Relationship between monthly CO$_2$ emission and N$_2$O emission from April to October.
Table 1 Correlation coefficients of the relationships between CO$_2$ or N$_2$O fluxes and soil physical and chemical properties.

<table>
<thead>
<tr>
<th>Year</th>
<th>Soil temperature $^\circ$C</th>
<th>WFPS %</th>
<th>NH$_4^+$-N (H$_2$O) mg N kg$^{-1}$</th>
<th>NH$_4^+$-N (KCl) mg N kg$^{-1}$</th>
<th>NO$_3^-$-N mg N kg$^{-1}$</th>
<th>WSOC mg C kg$^{-1}$</th>
<th>CO$_2$ flux mg C m$^{-2}$ h$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$n$</td>
<td>$R$</td>
<td>$n$</td>
<td>$R$</td>
<td>$n$</td>
<td>$R$</td>
<td>$n$</td>
</tr>
<tr>
<td>2000</td>
<td>19</td>
<td>0.54*</td>
<td>19</td>
<td>-0.17</td>
<td>19</td>
<td>-0.25</td>
<td>ND</td>
</tr>
<tr>
<td>2002</td>
<td>51</td>
<td>0.37*</td>
<td>43</td>
<td>0.09</td>
<td>33</td>
<td>-0.47**</td>
<td>ND</td>
</tr>
<tr>
<td>2003</td>
<td>34</td>
<td>0.68**</td>
<td>30</td>
<td>-0.51**</td>
<td>13</td>
<td>0.56*</td>
<td>ND</td>
</tr>
<tr>
<td>2004</td>
<td>26</td>
<td>0.68**</td>
<td>24</td>
<td>-0.34</td>
<td>18</td>
<td>-0.35</td>
<td>ND</td>
</tr>
<tr>
<td>2005</td>
<td>24</td>
<td>0.35</td>
<td>24</td>
<td>0.09</td>
<td>24</td>
<td>-0.37</td>
<td>ND</td>
</tr>
<tr>
<td>2006</td>
<td>27</td>
<td>0.31</td>
<td>29</td>
<td>0.22</td>
<td>29</td>
<td>-0.05</td>
<td>29</td>
</tr>
<tr>
<td>2007</td>
<td>25</td>
<td>0.13</td>
<td>21</td>
<td>-0.11</td>
<td>ND</td>
<td>ND</td>
<td>21</td>
</tr>
</tbody>
</table>

| Year  | N$_2$O flux (mg N m$^{-2}$ h$^{-1}$) | $n$ | $R$ | $n$ | $R$ | $n$ | $R$ | $n$ | $R$ | $n$ | $R$ |
|-------|-------------------------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| 2000  | 22                                  | 0.54**| 19  | -0.19 | 22  | -0.23 | ND  | ND  | 22  | 0.68**| ND  | ND  | 23  | 0.59**|
| 2002  | 51                                  | 0.26 | 43  | -0.11 | 33  | -0.30 | ND  | ND  | 33  | 0.04  | 33  | 0.08  | 60  | 0.74**|
| 2003  | 34                                  | 0.25 | 30  | -0.42*| 13  | 0.52  | ND  | ND  | 13  | 0.36  | 13  | -0.34 | 39  | 0.51**|
| 2004  | 26                                  | 0.50**| 24  | -0.32 | 18  | -0.36 | ND  | ND  | 18  | 0.78**| 18  | 0.03  | 29  | 0.71**|
| 2005  | 24                                  | 0.19 | 24  | 0.28  | 24  | -0.28 | ND  | ND  | 24  | 0.14  | 22  | 0.49* | 28  | 0.79**|
| 2006  | 28                                  | 0.21 | 29  | 0.09  | 29  | -0.01 | 29  | 0.10 | 28  | 0.23  | 29  | -0.10 | 32  | 0.55**|
| 2007  | 24                                  | -0.11| 21  | 0.23  | ND  | ND  | 21  | -0.20 | 22  | -0.37 | 22  | -0.37 | 25  | 0.27  |

* $P < 0.05$, ** $P < 0.01$
Table 2 Annual CO$_2$ and N$_2$O emissions from unfertilized bare soil in an onion field in Mikasa

<table>
<thead>
<tr>
<th>Year</th>
<th>Annual precipitation mm yr$^{-1}$</th>
<th>Mean annual air temperature $^\circ$C</th>
<th>Period day</th>
<th>CO$_2$ emission Mg C ha$^{-1}$ yr$^{-1}$</th>
<th>Uncertainty %</th>
<th>$Q_{10}$</th>
<th>N$_2$O emission kg N ha$^{-1}$ yr$^{-1}$</th>
<th>Uncertainty %</th>
</tr>
</thead>
<tbody>
<tr>
<td>2000</td>
<td>1576</td>
<td>7.7</td>
<td>302</td>
<td>3.32 a</td>
<td>14.2</td>
<td>2.38</td>
<td>4.60 bc</td>
<td>18.8</td>
</tr>
<tr>
<td>2002</td>
<td>1187</td>
<td>7.7</td>
<td>251</td>
<td>2.42 abc</td>
<td>16.9</td>
<td>2.03</td>
<td>3.94 bc</td>
<td>23.6</td>
</tr>
<tr>
<td>2003</td>
<td>986</td>
<td>7.4</td>
<td>318</td>
<td>2.04 c</td>
<td>20.7</td>
<td>1.80</td>
<td>2.01 c</td>
<td>74.0</td>
</tr>
<tr>
<td>2004</td>
<td>1294</td>
<td>8.4</td>
<td>280</td>
<td>2.48 bc</td>
<td>13.2</td>
<td>1.73</td>
<td>7.34 b</td>
<td>15.0</td>
</tr>
<tr>
<td>2005</td>
<td>1398</td>
<td>7.5</td>
<td>297</td>
<td>2.95 ab</td>
<td>21.2</td>
<td>1.53</td>
<td>12.1 a</td>
<td>19.0</td>
</tr>
<tr>
<td>2006</td>
<td>1124</td>
<td>7.8</td>
<td>271</td>
<td>2.25 bc</td>
<td>22.8</td>
<td>1.38</td>
<td>1.62 c</td>
<td>106</td>
</tr>
<tr>
<td>2007</td>
<td>1015</td>
<td>8.2</td>
<td>256</td>
<td>2.20 bc</td>
<td>58.9</td>
<td>1.11</td>
<td>2.56 c</td>
<td>153</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td></td>
<td>282</td>
<td>2.53 bc</td>
<td>24.0</td>
<td>1.71</td>
<td>4.88</td>
<td>58.5</td>
</tr>
</tbody>
</table>

Values within the same column differs significantly ($P < 0.05$)