



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
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17 5 Lowland soil in Mikasa, Hokkaido, Japan
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22 7 Running head: Variation in CO₂ and N₂O emission from bare soil
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32 **Abstract**

33 Soil is one of the important sources of atmospheric carbon dioxide (CO₂) and
34 nitrous oxide (N₂O). Study of CO₂ and N₂O emission from bare soil may explain
35 the annual change of carbon (C) in soil organic matter (SOM) and help analyzing
36 the N₂O production from SOM. Therefore, CO₂ and N₂O emissions associated
37 with the decomposition of SOM from bare soil are important factors for
38 assessing the C budget and N₂O emission in agricultural field. We conducted a
39 study over seven years to assess the controlling factors of CO₂ and N₂O
40 emissions from unplanted and unfertilized soil in Mikasa, Hokkaido, Japan.

41 Carbon dioxide flux increased in the summer, and there were significant
42 positive correlations between the CO₂ flux and soil temperature in the first four
43 years. However, apparent relationships between CO₂ flux and WFPS, soil NH₄
44 and NO₃ concentrations were not observed. The slope of monthly CO₂ emission
45 against mean monthly temperature was positively correlated with monthly
46 precipitation. These results suggest that response of CO₂ production by increase
47 in soil temperature becomes more sensitive in wet soils. The average CO₂
48 emission during the study period was 2.53 Mg C ha⁻¹ yr⁻¹, and uncertainty of the
49 annual CO₂ emission was 24 %. Annual precipitation explained the yearly

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6 50 variation (CO_2 emission [$\text{Mg C ha}^{-1} \text{ yr}^{-1}$] = $0.0021 \times \text{annual precipitation [mm yr}^{-1}] -$
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9 51 0.0499 , $R=0.976$, $P<0.001$).

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11 Nitrous oxide flux increased from July to October, and was positively
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14 53 correlated with CO_2 flux. Based on the ratio of $\text{N}_2\text{O-N:NO-N}$ of fluxes, N_2O
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17 54 appeared to be the main product of denitrification. The average N_2O emission in
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20 55 the study period was $4.88 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, and uncertainty of annual N_2O emission
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22 56 was 58.5 %. Strong relationships between the monthly emissions of CO_2 and
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25 57 N_2O suggest that N_2O production by denitrification is strongly affected by SOM
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28 58 decomposition. Unlike the CO_2 emission, the relationship between N_2O emission
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31 59 and precipitation was not observed because of the multiple pathways of
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34 60 nitrification and denitrification for N_2O production induced by SOM
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37 61 decomposition.
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39 63 **Key words:** carbon dioxide, denitrification, nitrous oxide, soil organic matter
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41 64 decomposition, temporal variation.
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6 66 **INTRODUCTION**
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9 67 The contributions of atmospheric carbon dioxide (CO₂) and nitrous oxide (N₂O)
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11 68 to global warming are reported to be 60% and 6%, respectively. Since the
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13
14 69 Industrial Revolution, concentrations of these greenhouse gases in the
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17 70 atmosphere have increased at a rate of 1.4 ppm yr⁻¹ and 0.8 ppb yr⁻¹,
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19 71 respectively (Intergovernmental Panel on Climate Change (IPCC) 2007) . The
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22 72 emission of carbon (C) that has originated due to the change in land use from
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25 73 1980 to 1989 contributes 24% of the global annual CO₂ emission. Similarly, the
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27 74 emission of N₂O originating from agricultural fields in 1989 contributed 24% of
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29
30 75 the total global N₂O emission (Mosier *et al.* 1998). Recently, the annual N₂O
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32 76 emissions from fertilized cropland and grassland at a global level were estimated
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35 77 at 3.3 and 0.8 Tg N yr⁻¹, respectively (Stehfest & Bouwman 2006).
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38 78 Some studies have reported that there is a reduction in C from agricultural
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40 79 soils because micro-organisms decompose soil organic matter (SOM) and emit
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43 80 CO₂ (Hu *et al.* 2004; Koga *et al.* 2006; Koizumi *et al.* 1993; Mu *et al.* 2006, 2008;
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46 81 Shimizu *et al.* 2009). The reduction of C from soil has been estimated by a
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48 82 difference in soil C over 20 years in a 0-30 cm soil surface (IPCC 2006; Paustian
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51 83 *et al.* 1997). In recent years, the global warming potential (GWP) has been used
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6 84 to evaluate the effect of agricultural activities on global warming (Chu *et al.* 2007;
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9 85 Jones *et al.* 2006; Koga *et al.* 2006; Mosier *et al.* 2005, Mu *et al.* 2006;
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11 86 Robertson & Grace 2004; Six *et al.* 2004). Therefore, it is necessary to estimate
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14 87 the annual reduction of soil C in order to evaluate the annual effect of agricultural
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17 88 activities on global warming. However, annual reduction of soil C is difficult to be
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20 89 detected from the investigation of annual change in the amount of soil C,
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22 90 because soil C is usually very large compared to C emission from the soil. In
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24 91 agricultural fields, decomposition of SOM can be measured as the CO₂ emission
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27 92 from bare fields in which root respiration is excluded (Hanson *et al.* 2000; Hu *et*
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30 93 *al.* 2004; Mu *et al.* 2006, 2008; Shimizu *et al.* 2009; Subke *et al.* 2006). Since the
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32 94 decomposition of SOM is resulted from microbial activities, it is influenced by
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35 95 chemical and physical conditions such as soil temperature, water conditions, pH,
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38 96 and so on. Due to the spatial variability of those factors (Yanai *et al.* 2003), it is
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41 97 difficult to investigate the factors affecting the decomposition of SOM at the field
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43 98 level. Understanding the factors affecting the decomposition of SOM, quantifying
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46 99 the annual CO₂ emission from bare fields, and clarifying factors affecting the
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49 100 temporal variations in annual CO₂ emissions from an unfertilized bare soil are
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51 101 important to estimate the loss of C from soil. Those understandings will provide
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6 102 beneficial information to the study of the effect of agricultural activities on C cycle
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9 103 or global warming at a long-term span such as eco-balance or life cycle analysis
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12 104 (Koga *et al.* 2006; Kimura *et al.* 2007).

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14 105 Nitrous oxide is produced by nitrification and denitrification processes in the
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17 106 soil. Therefore, N₂O emission is affected by the soil's chemical and physical
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20 107 conditions (Bremner 1997; Colbourn & Dowdell 1984; Mosier 1998; Stehfest &
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22 108 Bouwman 2006). Because ammonium (NH₄⁺) is used in the nitrification process
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24
25 109 and nitrate (NO₃⁻) in the denitrification process, the N₂O emission from
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28 110 agricultural fields is usually in proportion to the N application rate (Bouwman
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30 111 1996). Based on this, methods of estimating N₂O emissions from agricultural
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33 112 fields have been proposed (IPCC 2006). In Tier 1 and 2 in the IPCC Guidelines
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36 113 for National Greenhouse Gas Inventories (IPCC 2006), the emission factor (EF)
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38 114 is the ratio of N-induced N₂O emission, which is a difference between N₂O
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41 115 emissions from N-fertilized and unfertilized field, divided by the amount of
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44 116 applied N fertilizer. However, to calculate the original EF values, the N₂O
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47 117 emission originating from soil organic N should be considered, because N₂O
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50 118 emission from the soil in agricultural fields is usually composed not only of N₂O
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53 119 emissions originating from the applied N, but also of N₂O emissions induced by
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7 120 decomposition of SOM. The N₂O emission from bare fields is regarded as the
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9 121 emission originating from the soil organic N. There are several studies that have
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11 122 reported annual N₂O emissions from unfertilized bare fields (Akiyama *et al.*
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14 123 2006; Clayton *et al.* 1997; van Groenigen *et al.* 2004; Kamp *et al.* 1998; Koga *et*
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16
17 124 *al.* 2004; Zou *et al.* 2005). In addition, a temporal variation in N₂O emission from
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19 125 agricultural fields has been reported by several studies (Drury *et al.* 2006; Kusa
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22 126 *et al.* 2002; Takakai *et al.* 2006; Zou *et al.* 2005). In any case, however, the
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24 127 reported studies have not focused on the susceptible factors for N₂O flux or
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27 128 annual N₂O emission from SOM. There are few studies focusing on analysis of
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30 129 mechanisms of N₂O production and emission induced by SOM decomposition in
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33 130 agricultural bare fields at a long-term span. The knowledge about the controlling
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35 131 factors of N₂O flux and annual emissions will be also useful for the study of the
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38 132 effect of agricultural activities on C cycle or global warming in a long run.

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40 133 The objective of this study was to clarify the controlling factors for CO₂ and
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43 134 N₂O emissions from agricultural bare soil based on seven years of monitoring in
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46 135 central Hokkaido, Japan.

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51 137 **MATERIALS AND METHODS**
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6 138 **Site description**
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9 139 This study was conducted in Mikasa, central Hokkaido, Japan (43°14.4 'N,
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11 140 141°50 'E). The soil type is Gray Lowland soil (Gleysol; FAO/UNESCO). The
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13
14 141 mean annual temperature of the study site is 7.5°C and the mean annual
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17 142 precipitation is 1164 mm, of which 32% is in the form of snow. The soil does not
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19 143 freeze during the winter. Temperature usually increases in August and
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22 144 precipitation increases from July to October (Fig. 1). At a depth of 0–10 cm, the
23
24 145 soil pH (H₂O) was 5.8 and the cation exchange capacity was 25.5 cmolc kg⁻¹.
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27 146 The soil was comprised of sand (12.1%), silt (51.2%) and clay (36.7%).
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30 147 Concentrations of soil C and N were 32.1 and 2.8 g kg⁻¹, respectively (Toma &
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32 148 Hatano 2007) and the C:N ratio was 11.5 at a depth of 0–10 cm in 1996. Soil C
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34 149 density was recorded as 108 Mg C ha⁻¹ at a 0-30 cm depth.
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40 151 **Experimental design**
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43 152 We set up an unfertilized bare plot (5 m × 8 m) inside a 2-ha onion field owned
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45 153 by a farmer in Mikasa in 1999 and conducted monitoring in 2000, from 2002 to
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48 154 2005, and in 2007. In 2006, the unfertilized bare plot was shifted to another
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51 155 location in the same field by the farmer due to unavoidable circumstances.
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7 156 Conventional management practices for onion cultivation were carried out on the
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9 157 studied plot every year, although chemical and organic fertilizers or residues
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11 158 were not applied. Most of weed was removed from the monitoring plot regularly.
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14 159 Onion is usually cultivated from May to September in Mikasa. The soil is usually
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16
17 160 tilled before fertilization and plowed up to a depth of 15 cm at the end of April.
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19 161 Onion roots are cut for harvest from mid to late August. The onion harvest is
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22 162 followed by plowing, which is carried out to incorporate the residues into the soil
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25 163 from the end of September to early October.
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30 165 **Measurement of CO₂, N₂O, and NO fluxes**

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32 166 Fluxes of CO₂, N₂O, and NO were measured by a closed chamber method
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35 167 (three or four replications) using two types of cylindrical stainless steel chambers
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38 168 from 10:00 am to 2:00 pm. In 2000, chambers of 30 cm in diameter and 35 cm in
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41 169 height were used (Kusa *et al.* 2002, 2006), whereas chambers of 20 cm in
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44 170 diameter and 25 cm in height were used from 2002 to 2007. The cover of the
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46
47 171 chamber was made of acryl and was equipped with a sample collector, a
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50 172 pressure regulating bag and a Tedlar bag (0.5 L). In 2000 and from 2002 to 2004,
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53 173 we inserted the chambers directly into the soil to a 2 cm depth and started
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6 174 making measurements after 15 min. However, in 2005, 2006, and 2007, we used
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8
9 175 a chamber-base made of stainless steel with a diameter of 20 cm. The upper
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11 176 part of the chamber-base had a slight depression, which was filled with water to
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14 177 seal it during the measurement (Toma & Hatano 2007; Toma *et al.* 2007). The
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17 178 base was kept on the ground, except during plowing and was reset one day
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19 179 before the next sampling.
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22 180 Gas samples were taken at time 0 min from inside the chamber and 6 min. (for
23
24 181 CO₂) or 15 min. (for N₂O and NO) after closing the chamber (Nakano *et al.* 2004;
25
26
27 182 Toma & Hatano 2007). Using a 25 mL syringe, gas sample was taken ten times
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30 183 (total volume of 250 mL), and the gas sample was injected into a Tedlar bag (0.5
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32 184 L). The bags were then brought to the laboratory and 20 mL of each gas sample
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34
35 185 was immediately transferred into glass vials (10 mL). From the samples of the
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37
38 186 bags, CO₂ and NO were analyzed using an Infrared CO₂ Analyzer (Model
39
40 187 ZFP5YA3I, Fuji Electric, Tokyo, Japan) and a Chemoluminescence Nitrogen
41
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43 188 Oxide Analyzer (Model 265P, Kimoto Electric, Osaka, Japan), respectively. From
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46 189 the samples of the vials, N₂O was analyzed with an ECD Gas Chromatograph
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48 190 (Model GC-14B, Shimadzu, Kyoto, Japan).
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51 191 The gas fluxes were calculated using the following equation:
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$$192 \quad F = \rho \times (V/A) \times (\Delta c/\Delta t) \times [273/(273 + T)] \times (P/760)$$

193 where, F is the flux (mg N or C m⁻² h⁻¹), ρ is the gas density (ρ_{N₂O-N} = 1.259
194 × 10⁶, ρ_{CO₂-C} = 0.536 × 10⁶ and ρ_{NO-N} = 0.625 × 10⁶ mg m⁻³), V is the volume
195 of the chamber (m³), A is the area of the chamber (m²), Δc/Δt is the ratio of
196 change in the gas concentration inside the chamber (10⁻⁶ m³ m⁻³ h⁻¹), T is the
197 air temperature inside the chamber (°C), and P is the air pressure (mm Hg; see
198 Kusa *et al.* 2002; Toma & Hatano 2007).

199 A positive value of F indicates gas emission from the soil to the atmosphere,
200 while the negative value indicates gas uptake by the soil from the atmosphere.
201 Considering the machinery precision, N₂O flux values within the range from -6 to
202 6 μg N m⁻² h⁻¹, and CO₂ flux values from -3.7 to 3.7 mg C m⁻¹ h⁻¹ and NO flux
203 values from -0.2 to 0.2 μg N m⁻² h⁻¹ were regarded as 0 μg N or C m⁻² h⁻¹. The
204 annual CO₂ and N₂O emissions were calculated assuming linear changes
205 between two sampling occasions.

206 The value of Q₁₀, which is a relative increase of CO₂ flux for 10°C change in
207 soil temperature, was calculated based on the Arrhenius equation and following
208 equation (Hu *et al.* 2004): $k = A \times \exp(-E/RT) \rightarrow \ln k = \ln A - E/RT$

$$Q_{10} = \frac{A \times \exp\{-E / R(T + 10)\}}{A \times \exp(-E / RT)}$$

209 where, k is the CO_2 flux, A is a constant, E is the activation energy, R is the
210 universal gas constant ($8.3144 \text{ J mol}^{-1} \text{ K}$), and T is the absolute temperature (K).
211 The logarithm of the CO_2 flux and the reciprocal of absolute temperature of soil
212 were plotted, and E was determined from the slope. The value of Q_{10} was
213 calculated by using $T = 283 \text{ (K)}$, because mean monthly air temperatures in this
214 study site ranged from 10°C to 20°C (Fig. 1c and d) .
215

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

218 The soil temperature at a 5 cm depth was measured five times at the same time
219 of measuring CO_2 , N_2O , and NO fluxes. Disturbed soil samples (0-5 cm depth)
220 were taken at five replications and were combined for one composite sample.
221 Soil solutions were extracted by using distilled water through the study period
222 and $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$ and water soluble organic carbon (WSOC) concentrations
223 were analyzed by colorimetry with indophenol-blue, ion chromatography (QIC
224 Analyzer, Dionex, Osaka, Japan) and total organic carbon analyzer (Model
225 TOC-5000A, Shimadzu, Kyoto, Japan), respectively. In 2007, soil $\text{NH}_4^+\text{-N}$

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7 226 concentration extracted by distilled water was not measured, and in 2006 and
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9 227 2007, soil NH_4^+ -N concentration extracted by 2M KCl were analyzed. The
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11 228 WSOC was not measured in 2000. The undisturbed soil samples were collected
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14 229 by using a steel cylinder (100 mL) at three replicates and the water-filled pore
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17 230 space (WFPS) was measured. The daily or annual meteorological data were
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19 231 obtained from the local Iwamizawa Weather Station (43°12.6 'N, 141°47.3 'E).

22 232 After plowing, all samples were collected three times a week from May to June,
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24 233 three times a month from July to August and once or twice a month during the
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27 234 snow-cover season. During the period from snowmelt to onion cultivation and
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30 235 from mid-October to snow cover, sampling was conducted two or three times per
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32 236 month.

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37 238 **Statistical analyses**

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40 239 Relationships between CO_2 and N_2O fluxes and soil physical and chemical
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42 240 properties, or annual CO_2 and N_2O emissions and of mean annual air
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45 241 temperature or annual precipitation were analyzed by the linear or quadratic
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48 242 curve regression. Variation in annual CO_2 or N_2O emissions among the study
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51 243 period was evaluated by Tukey-Kramer test. The least significant difference test
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7 244 was used to determine significant differences ($P < 0.05$).

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9 245 Uncertainties were calculated using the following equation:

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

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14 247 The two-sided 95% confidence interval of CO₂ and N₂O emissions were
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16
17 248 calculated by using the following equation:

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

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21
22 250 where, d.f. is the degree of freedom, and $t(\text{d.f.}, 0.05)$ is the t value at 5%
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25 251 significance level with two-sided alternative.

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31 32 254 **RESULTS**

33 34 255 **Seasonal variation in CO₂ and N₂O fluxes**

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37 256 The CO₂ flux peaked from August to October (Fig. 2a, 3a), whereas the N₂O flux
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40 257 increased from the end of July to October in all years (Fig. 2b, 3b). Soil
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43 258 temperature increased in August and this trend was similar to that of air
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46 259 temperature (Fig. 4a,f). The WFPS was high in early April and November, and
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49 260 low in August (Fig. 4b,g). From August to October, the WFPS was as low as
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51 261 around 50%. Soil NH₄⁺-N concentrations extracted by distilled water ranged from
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6 262 0 to 7.6 mg N kg⁻¹ each year and did not exhibit any consistent seasonal change
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9 263 (Fig. 4c,h). Although NH₄⁺-N concentration extracted by 2M KCl in 2006 and
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11 264 2007 did not show a distinct seasonal change (Fig. 4i), the range of
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14 265 concentrations (1.67-11.3 mg N kg⁻¹ in 2006, 0.84-29.9 mg N kg⁻¹) was higher
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17 266 than the values obtained through extraction by water. Soil NO₃⁻-N concentrations
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19 267 increased from August to September each year (Fig. 4d,j). Fluctuation in soil
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21
22 268 WSOC concentrations was small throughout the year (Fig. 4e,k).

23
24 269 Correlation coefficients between the CO₂ or N₂O fluxes and soil physical and
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26
27 270 chemical properties are given in Table 1. The CO₂ flux was significantly and
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29
30 271 positively correlated with soil temperature from 2000 to 2004 (Table 1), and
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32 272 reached its maximum at 48.8% of WFPS (Fig. 5a). Although the N₂O flux was
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34
35 273 significantly and positively correlated with soil temperature in 2000 and 2004,
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37
38 274 N₂O flux was significantly and positively correlated with CO₂ flux from 2000 to
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40
41 275 2006. Carbon dioxide and N₂O fluxes were significantly correlated with soil
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43 276 NO₃⁻-N concentration, and N₂O flux was significantly correlated with soil
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46 277 temperature in 2000 and 2004. However, no significant correlation was observed
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49 278 between CO₂ or N₂O fluxes and soil chemical or physical properties throughout
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51 279 the study period. There were no significant correlations between N₂O flux and
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6 280 WFPS during the study period except for 2003, but N₂O showed high fluxes
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9 281 when WFPS ranged from 30 to 60 % and reached its maximum at 48.4% (Fig.
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11 282 5b).

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14 283 The relationship between N₂O flux and N₂O-N:NO-N ratio is given in Fig. 6.

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17 284 The N₂O flux increased with an increase in N₂O-N:NO-N ratio, with 18 out of 49
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19 285 samples of the N₂O flux over 0.079 mg N m⁻² h⁻¹, which resulted in the ratio of
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22 286 N₂O-N:NO-N over 100. The average N₂O flux was 0.079 mg N m⁻² h⁻¹.

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25 287 Therefore, about 37% of the N₂O fluxes were higher than the average value.

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28 289 **Cumulative CO₂ and N₂O emissions**

29
30 290 Table 2 shows the annual CO₂ and N₂O emissions and Q₁₀ values. The annual

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33 291 CO₂ emission ranged from 2.04 to 3.32 Mg C ha⁻¹ yr⁻¹, and were significantly

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36 292 correlated with annual precipitation (Fig. 7, $y = 0.0021x - 0.0499$, $R = 0.98$, $P <$

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39 293 0.01). The average CO₂ emission during the study period was 2.53 Mg C ha⁻¹

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42 294 yr⁻¹, and uncertainty of annual CO₂ emission was 24 %. The Q₁₀ values ranged

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45 295 from 1.11 to 2.38. There was no significant correlation between Q₁₀ value and

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48 296 annual precipitation ($y = 0.0012x + 0.1918$, $R = 0.56$, $P = 0.11$). However, the

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51 297 slope of the monthly CO₂ emission against mean monthly temperature from April

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7 298 to October was significantly correlated with precipitation from April to October
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9 299 (Fig. 8, $y = 0.0688x - 27.67$, $R = 0.90$, $P < 0.01$). Carbon dioxide flux in this study
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11 300 was measured only during the daytime, and annual CO₂ emission was
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14 301 calculated assuming linear changes between two sampling occasions. This
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17 302 indicated that the decrease in CO₂ flux during the night time, when temperature
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19 303 decreased, was not considered in calculating the annual CO₂ emission from
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21
22 304 2000 to 2004. The difference in maximum and minimum air temperature from
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24
25 305 July to September, which was the period of high air temperature, was below
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27 306 10°C (8.8°C). In addition, the average Q₁₀ value from 2000 to 2004 was 1.99
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30 307 (Table 2). If we assume that the CO₂ flux during the maximum daily air
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32 308 temperature is 1.99 times higher than the CO₂ flux during minimum daily air
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35 309 temperature, the measured annual CO₂ emission would be 1.33 times higher
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38 310 than the annual CO₂ emission that was calculated by considering the daily
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41 311 change in the CO₂ flux.

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43 312 The annual N₂O emission ranged from 1.62 to 12.1 kg N ha⁻¹ yr⁻¹. The average
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45 313 N₂O emission during the study period was 4.88 kg N ha⁻¹ yr⁻¹, and uncertainty of
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48 314 the annual N₂O emission was 58.5 %. Uncertainty of yearly variation of CO₂ and
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51 315 N₂O emissions were 17.9 and 76.1 %, respectively. Although correlations
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7 316 between N₂O emission and annual air temperature or precipitation were not
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9 317 significant, N₂O emission increased with an increase in CO₂ emission (Fig. 9).
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11 318 Significant differences in annual CO₂ and N₂O emissions among the study years
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14 319 indicate yearly variation of CO₂ and N₂O emission in the study site (Table 2).
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18 19 321 **DISCUSSION**

20 21 22 322 **Key factors determining the CO₂ flux in bare soil**

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25 323 Several studies reported that CO₂ flux from soil showed an exponential increase
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27 324 with an increase in soil temperature (Boone *et al.* 1998; Hu *et al.* 2001; Jones *et*
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30 325 *al.* 2006; Schindlbacher *et al.* 2009). In this study, there were significant positive
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32 326 correlations between CO₂ flux and soil temperature (in 2000, 2002-2004) (Table
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35 327 1). On the other hand, there was no significant correlation between CO₂ flux and
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38 328 WFPS except in 2003 (Table 1). Gulledge and Schimel (1998) reported that
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40 329 microbial respiration was directly proportional to the water holding capacity
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43 330 (10-60%). Moreover, Linn and Doran (1984) and Gulledge and Schimel (1998)
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46 331 reported that the maximum CO₂ production by microorganisms was at 60%
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49 332 WFPS. In our study, CO₂ flux peaked at 49.1% of WFPS. Therefore, soil
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51 333 moisture might not have influence on the production or emission of CO₂ linearly,
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7 334 and there might be an appropriate condition of soil moisture for CO₂ production
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9 335 relating to the ventilation of soil. On the other hand, significant positive
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11 336 correlations between annual CO₂ emission and annual precipitation and
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14 337 between the slope of monthly CO₂ emission against mean monthly temperature
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17 338 from April to October and precipitation from April to October suggested that the
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19 339 production of CO₂ was greater in wet conditions even if temperatures were
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22 340 similar (Fig. 7, 8). Therefore, CO₂ production increased with an increase in soil
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25 341 temperature, but CO₂ production at the same temperature was more enhanced
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27 342 in wet conditions. High moisture content prevents soil gas from diffusing from soil
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30 343 to the atmosphere. This means that even if CO₂ was produced in soil in
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32 344 appropriate condition for CO₂ production, CO₂ might not necessarily be diffused
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35 345 at that time. There could be possibilities of high CO₂ flux to be detected during
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37 346 the season of high precipitation. Difference in timing between CO₂ production
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40 347 and diffusion from soil to the atmosphere could possibly be explained by a lack
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43 348 of correlation between CO₂ flux and WFPS and by a significant positive
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46 349 correlation between annual CO₂ emission and precipitation (Table 1, Fig. 7).
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51 **Key factors determining the N₂O flux in bare soil**
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7 352 Because N₂O is mainly produced by the processes of nitrification and
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9 353 denitrification in soil (Tiedje 1994), N₂O flux is often affected by soil NH₄⁺-N or
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11 354 NO₃⁻-N concentration (Livesley *et al.* 2009; Thornton & Valente 1996; Toma *et al.*
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13
14 355 2007). However, no apparent correlation between N₂O flux and soil NH₄⁺-N or
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16 356 NO₃⁻-N concentration was observed in our study. Instead, we observed
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18
19 357 significant positive correlations between N₂O and CO₂ fluxes in all years, except
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22 358 for 2007 (Table 1). Similar positive correlations between N₂O and CO₂ fluxes
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25 359 were observed when crop residues were mixed with surface soil (Huang *et al.*
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27 360 2004; Toma & Hatano 2007). In this study, the substrates for the nitrification and
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29
30 361 denitrification originated from SOM because no fertilizer was applied in the
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33 362 experimental plot, suggesting that N₂O production was strongly related with
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35 363 SOM decomposition. Therefore, one of the factors controlling the production of
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38 364 N₂O could be the supply of C and inorganic N from SOM decomposition. Some
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41 365 studies reported that soil temperature is one of the factors for N₂O production
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43 366 (Kamp *et al.* 1998; Mori *et al.* 2005; Smith *et al.* 1998; Tokuda & Hayatsu 2004).
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46 367 Since the CO₂ flux increased with an increase in soil temperature in this study
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49 368 (Table 1), soil temperature might not affect only the activity of N₂O production but
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51 369 also the supply of substrates for N₂O production. Bouwman (1990) summarized
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6 370 the results presented by Anderson and Levine (1986) and Lipchults *et al.* (1981)
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8
9 371 and reported that a value of $N_2O-N:NO-N$ ratio below 1.0 indicates that N_2O was
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11 372 produced in the soil mainly by a nitrification process, while a value above 100
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14 373 indicates that denitrification was the dominant process for N_2O production. In our
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17 374 study, About 37% of the N_2O fluxes that were higher than the average value,
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19 375 therefore, occurred when there was a condition of $N_2O-N:NO-N$ ratio to be above
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22 376 100. Generally, an increase in soil moisture contributes to N_2O production by
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24 377 denitrification (Linn & Doran 1984). It was reported that a WFPS of 50-60%
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27 378 would be suitable for the decomposition of soil organic matter and that these
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30 379 conditions accelerated denitrification due to the consumption of oxygen (Maag &
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32 380 Vinther 1999). Moreover, WFPS of 50-60% was the range in which N_2O was
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35 381 mainly produced by the denitrification process (Davidson *et al.* 2000). In our
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38 382 study, WFPS increased from about 40 to 60% from the end of July to early
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40 383 October, and N_2O flux was highest when the WFPS was at 46.5% (Fig. 4, 5).
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43 384 Sawamoto and Hatano (2000) reported that an increase in N_2O production in
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46 385 autumn might be caused by nitrate transportation and diffusion inside soil
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49 386 aggregates after rainfall and development of denitrification areas inside the
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51 387 aggregates of well-structured soil in the same field of our study site. Therefore,
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7 388 there is a possibility of aerobic and anaerobic conditions occurring
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9 389 simultaneously inside the aggregates and inter-aggregate pores in the
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11 390 well-structured soil in this study. This would suggest that high N₂O fluxes were
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14 391 induced not only by denitrification but also by nitrification. Nitrous oxide was the
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17 392 main product of the denitrification process in this study.
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19 393 Nitrous oxide fluxes were significantly correlated with CO₂ flux, and production
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22 394 of CO₂ was affected by soil temperature and precipitation. However, a significant
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25 395 correlation between N₂O flux or annual N₂O emission and WFPS or annual
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28 396 precipitation could not be found. This might be due to a complex process of N₂O
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30 397 production in soil. In this study, the source of inorganic N might be mainly SOM.
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32 398 Therefore, NH₄ and NO₃, which are substrates for nitrification and denitrification,
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35 399 could have been produced by mineralization and nitrification processes in soil. In
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37
38 400 the study of N₂O emission from organic fertilizer, similar correlations between
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41 401 N₂O and CO₂ emission were observed (Hayakawa *et al.* 2009). When N
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43 402 originates from organic matter, decomposition of organic matter is probably
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46 403 essential for producing N₂O. In any case, significant correlation between CO₂
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49 404 flux and N₂O flux or monthly emission of CO₂ and N₂O suggest that climate
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51 405 change, which will affect SOM decomposition, might greatly affect N₂O emission
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6 406 induced by SOM decomposition.
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11 408 **Annual CO₂ emission and its spatial and temporal variations**
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14 409 The annual CO₂ emission, associated with decomposition of SOM, ranged from
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17 410 2.04 to 3.32 Mg C ha⁻¹ yr⁻¹ (Table 2). The values of CO₂ emission from 2000 to
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19 411 2004, however, might have been overestimated about 1.33 times because daily
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22 412 variation in CO₂ flux was not considered. Therefore, if we consider the
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24
25 413 overestimation of CO₂ emission, annual CO₂ emission ranged from 1.11 to 2.95
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27 414 Mg C ha⁻¹ yr⁻¹. Mu *et al.* (2006) reported that CO₂ emitted from SOM in bare
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30 415 treatments in several types of land use (e.g. wheat, maize, onion, etc.) ranged
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32 416 from 3.01 to 5.68 Mg C ha⁻¹ yr⁻¹ in Hokkaido, Japan. Jacinthe *et al.* (2002)
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35 417 reported that 4.37 Mg C ha⁻¹ yr⁻¹ of CO₂ was emitted from a bare plot in Ohio,
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38 418 USA. Decomposition of SOM in the two studies might have been overestimated
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40 419 similar to our study because they used the trapezoidal rule to calculate annual
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43 420 CO₂ emission. Koizumi *et al.* (1994) reported that heterotrophic respiration
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46 421 ranged from 7.16 to 10.5 Mg C ha⁻¹ yr⁻¹ in three double-cropping
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48 422 agro-ecosystems (rice-barley, peanut-wheat, and dentcorn-italian ryegrass).
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51 423 They determined the SOM decomposition as the difference between total soil
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7 424 respiration and plant respiration by calculating soil and root respirations using
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9 425 continuous data of soil temperature and the correlations between soil
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11 426 temperature and soil or root respirations. Koga *et al.* (2006) reported average
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13 427 SOM decomposition at a 20-year time scale since 1981 as 1.04 to 1.34 Mg C
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15 428 ha⁻¹ yr⁻¹ from the difference in total soil C within a depth of 20 cm in Hokkaido,
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17 429 Japan. The decomposition rate of SOM reported by Koizumi *et al.* (1994) and
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19 429 Japan. The decomposition rate of SOM reported by Koizumi *et al.* (1994) and
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21 430 Koga *et al.* (2006) might not have been overestimated, and were larger and
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24
25 431 smaller than the values in this study, respectively. The reason of the difference in
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27 432 SOM decomposition may be due to the variation in location, cultivation system,
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29 433 and soil type of the study site. The mean annual temperature (13.1°C) of the
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31 434 study site reported by Koizumi *et al.* (1994) was higher than that in our study site.
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33 435 In addition, there were frequent tillage operations because of a double-cropping
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35 436 system. High temperature and frequency of plowing may have increased SOM
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37 437 decomposition in the study reported by Koizumi *et al.* (1994). On the other hand,
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39 438 the study site of Koga *et al.* (2006) was located in similar latitude (42°53 'N) as
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41 439 this study. Therefore, difference in SOM decomposition between our study and
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43 440 the study conducted by Koga *et al.* (2006) might be due to the differences in crop
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45 441 management tillage, and/or soil type. Mu *et al.* (2008) reported that CO₂
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6 442 emission from bare soil reached to its maximum at 60-70% of the sum of silt and
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9 443 clay content in soil. Moreover, Bellamy *et al.* (2005) reported that an amount of C
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11 444 during 25-year was decreased with an increase soil C content. In these studies,
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13
14 445 however, soil type of Andisols was not included. Further study about the
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17 446 difference in SOM decomposition in various soil types will be required.

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19 447 Carbon dioxide emission was measured at the same location in this study site
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22 448 from 2000 to 2005. During this period, average annual CO₂ emission was 2.84
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24 449 Mg C ha⁻¹ yr⁻¹, and 17 Mg C ha⁻¹ was estimated to be released from soil in 2000 -
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27 450 2005. Because mass of C in surface (0-30cm) soil was 108 Mg C ha⁻¹, C loss of
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30 451 17 Mg C ha⁻¹ corresponded to 15.7 % of surface soil C. We did not measure the
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33 452 change in soil C storage during the study period. Therefore actual value of
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36 453 reduction in soil C could not be shown in this study. Mass of soil C possibly could
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38 454 be detected, if soil C change was measured during this study. Bellamy *et al.*
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40 455 (2005), however, reported that loss of soil C was not detectable over 10 years
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43 456 when soil C content was lower than 50 g kg⁻¹. Detecting the change in soil C in
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46 457 this study site might be difficult. There are several ways of C input in agricultural
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49 458 fields such as rainfall, weed, and alga. But most of weed or alga did not exist
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51 459 during the study period. Carbon input from rainfall was not included in the IPCC
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6 460 guideline for calculating soil C change (IPCC 2006). Thus, there might be no
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9 461 major C inflow that supplemented soil C loss by SOM decomposition. Further
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11 462 study would be required for matching the reduction of soil C and CO₂ emission
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14 463 from unfertilized bare soil.

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17 464 A large variation in CO₂ fluxes and annual CO₂ emissions among replication
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19 465 were observed due to the spatial variation in CO₂ flux (Fig. 2a, 3a, and Table 2).
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21
22 466 The average uncertainty of annual CO₂ emission (24 %) was higher than that of
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24 467 annual CO₂ emission (17.9 %). The decomposition of SOM is generally affected
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26
27 468 by soil properties (Koizumi *et al.* 1993; Mu *et al.* 2008), method of cultivation
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30 469 (Koga *et al.* 2006), or land use practices (Mu *et al.* 2006). Mu *et al.* (2008)
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33 470 reported a significant correlation between CO₂ emission and clay and silt content
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35 471 in a bare field in the same district of our study site. In the study field, spatial
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38 472 variation in total C content, soil C:N ratio, and microbial biomass C were reported
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40 473 (Yanai *et al.* 2003). These factors possibly cause large uncertainty in annual CO₂
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43 474 emission. Using a larger chamber or taking more replication for CO₂ flux
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46 475 measurement might be required to improve an accuracy of annual CO₂
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48 476 emissions.

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51 477 Significant differences in annual CO₂ emissions from bare field during the
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7 478 study period show that there is a yearly variation in annual CO₂ emission that is
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9 479 induced by the SOM decomposition (Table 2). A large uncertainty (17.9%) of the
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11 480 average of annual CO₂ emission during the study period shows that continuous
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14 481 monitoring of CO₂ emission is required to determine the representative value of
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17 482 annual CO₂ emission. This yearly variation in annual CO₂ emission may suggest
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19 483 that climate will have a great impact on yearly CO₂ emission from SOM. Raich
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21
22 484 and Schlesinger (1992) reported that soil respiration, including root respiration,
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25 485 increased with an increase in annual precipitation or mean annual air
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27 486 temperature. Our study also showed that annual precipitation affect CO₂
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30 487 emission from mineralization of SOM in the study site.

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32 488 Previous studies reported that the effect of chemical fertilizer application on
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35 489 microbial respiration is negligible (Ginting *et al.* 2003; Hu *et al.* 2004; Jacinthe *et*
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38 490 *al.* 2002). Hence, CO₂ emissions obtained in this study could be an
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41 491 approximation of SOM decomposition in a fertilized field. Several studies
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44 492 reported that soil C decreased in agricultural fields with low levels of organic
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47 493 matter application (Ginting *et al.* 2003; Hu *et al.* 2004; Jacinthe *et al.* 2002; Mu *et*
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50 494 *al.* 2008; Shimizu *et al.* 2009). The loss of soil C is reported to have a greater
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60 495 influence on global warming than the emissions of N₂O or CH₄ (Jones *et al.*

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6 496 2006; Koga *et al.* 2006; Mosier *et al.* 2005; Mu *et al.* 2006). Although it is difficult
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9 497 to calculate an annual loss of C by measuring soil C reduction, it could possibly
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11 498 be done by measuring CO₂ emission from bare soil in an agricultural field.
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14 499 Assessing CO₂ emissions from bare fields, by calculating from the correlation
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17 500 between CO₂ emission and available environmental factors such as
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19 501 meteorological data or soil type, will be useful for cultivation management or
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22 502 planning of farm activities at a regional scale to mitigate C loss from soil.
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25 503

26 27 504 **Annual N₂O emission and its spatial and temporal variations**

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29
30 505 The annual N₂O emission in this study ranged from 1.62 to 12.1 kg N ha⁻¹ yr⁻¹
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32 506 (average 4.88 kg N ha⁻¹ yr⁻¹). These values are higher than most reported values
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35 507 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*
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37 508 *al.* 2004), 0.14-1.52 kg N ha⁻¹ yr⁻¹ (van Groenigen *et al.* 2004) or 0.36-0.14 kg N
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40 509 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
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43 510 ha⁻¹ yr⁻¹ (Zou *et al.* 2005). The difference in annual N₂O emission might be due to
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46 511 soil physical and chemical properties. Nitrogen content in soil may affect N₂O
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49 512 production because there is no any N source for a substrate of N₂O in bare soil
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51 513 except for N in SOM. Klemedtsson *et al.* (2005) reported at significant negative
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6 514 relationship between N₂O emission and soil C:N ratio in forested peat soil in
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9 515 Europe, indicating that N₂O emission increased with an increase in relative N to
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11 516 C in peat. Nitrogen contents of the soils in the study sites of Kamp *et al.* (1998)
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13 517 (1.7 g kg⁻¹) and Zou *et al.* (2005) (1.1 g kg⁻¹), in which N₂O emissions exceeded
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15
16 518 4.0 kg N ha⁻¹, were lower than the value of this study site (2.8 g kg⁻¹). Production
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19 519 of N₂O in poorly drained soil increases due to an anaerobic condition in soil (van
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21 520 Groenigen *et al.* 2004). Nitrous oxide emission reported by Koga *et al.* (2004)
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23
24 521 was lower than the value of this study even if N content in their study site
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27 522 (2.4-3.1 g kg⁻¹) was similar to that of our study. This could possibly be due to the
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29 523 drainage quality of soil. Soil type in the study site of Koga *et al.* (2004) is
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32 524 well-drained volcanic ash soil. Therefore, N concentration as a source of
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35 525 substrate for N₂O and drainage condition that may control aerobic- and
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38 526 anaerobic condition of soil might be indicator of the potential N₂O source from
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41 527 unfertilized bare field.

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43 528 Large variations in flux and annual emission of both N₂O and CO₂ were
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46 529 observed in our study (Fig. 1b, 2b and Table 2). There is considerable
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49 530 uncertainty in the amount of N₂O emission from unfertilized bare fields (Akiyama
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51 531 *et al.* 2006; Zou *et al.* 2005). Both CO₂ and N₂O emissions were influenced by
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6 532 soil properties (van Groenigen *et al.* 2004) and cultivation methods (Koga *et al.*

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9 533 2004). In this study site, high spatial variability (CV 217%) of N₂O flux was

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11
12 534 reported (Yanai *et al.* 2003). Yanai *et al.* (2003) also reported that soil organic

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14 535 matter and soil pH were main soil-related determining factors for N₂O flux.

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16
17 536 The temporal variation in N₂O was confirmed to be similar to that in some

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19 537 previous studies (Drury *et al.* 2006; Kusa *et al.* 2002; Takakai *et al.* 2006; Zou *et*

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22 538 *al.* 2005). In this study, neither N fertilizer nor crop residue was applied in the

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25 539 experimental plot from 1999 to 2005. Therefore, it is expected that the substrate

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27 540 required for production of N₂O had been reduced. However, the annual N₂O

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30 541 emission did not consistently decrease from 2000 to 2005, resulting in a large

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33 542 uncertainty of annual N₂O emission (Table 2). This suggests that the effect of

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35 543 climatic factors on N₂O emission might be greater than the reduction in substrate

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38 544 required for N₂O production. Annual N₂O emission tended to increase with an

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41 545 increase in annual precipitation. The relationship between annual N₂O emission

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43 546 and annual precipitation was not significant, whereas annual CO₂ emission was

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46 547 significantly correlated with annual precipitation (Fig. 7). The reason for this

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49 548 could be due to the fact that there is a difference in environmental factors that is

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51 549 required for mineralization, nitrification, and denitrification. In mineralization and

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7 550 nitrification processes, oxygen is required for oxidation of organic C or NH_4 .
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9 551 However, an anaerobic condition is required for denitrification because NO_3 is
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11 552 used as an electron capture (Bouwman and Boumans 2001). Since N_2O was
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14 553 produced through these three biological processes in soil, a correlation between
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17 554 annual N_2O emission and annual precipitation may not be significant.
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6 752 **Figure legends.**
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9 753 **Figure 1** Seasonal variations in monthly precipitation [a, b] and mean monthly
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11 754 air temperature [c, d].
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16 756 **Figure 2** Seasonal variations in CO₂ [a], N₂O [b], and NO [c] fluxes in 2000,
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18 757 2002, and 2003. Arrows indicate the timing of plowing.
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24 759 **Figure 3** Seasonal variations in CO₂ [a], N₂O [b], and NO [c] fluxes from 2004 to
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26 760 2007. Arrows indicate the timing of plowing.
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32 762 **Figure 4** Seasonal variations in soil temperature [a & f], WFPS [b & g], soil
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34 763 NH₄⁺-N [c, h & j], NO₃⁻-N [d & i], and WSOC [e & j] concentrations.
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40 765 **Figure 5** Relationships between CO₂ or N₂O fluxes and WFPS.
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45 767 **Figure 6** Relationship between N₂O flux and N₂O-N:NO-N ratio.
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51 769 **Figure 7** Relationship between CO₂ emission and annual precipitation.
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9 771 **Figure 8** Relationship between slope of the monthly CO₂ emission against mean

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11 772 monthly temperature and precipitation from April to October.
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17 774 **Figure 9** Relationship between monthly CO₂ emission and N₂O emission from

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19 775 April to October.
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For review

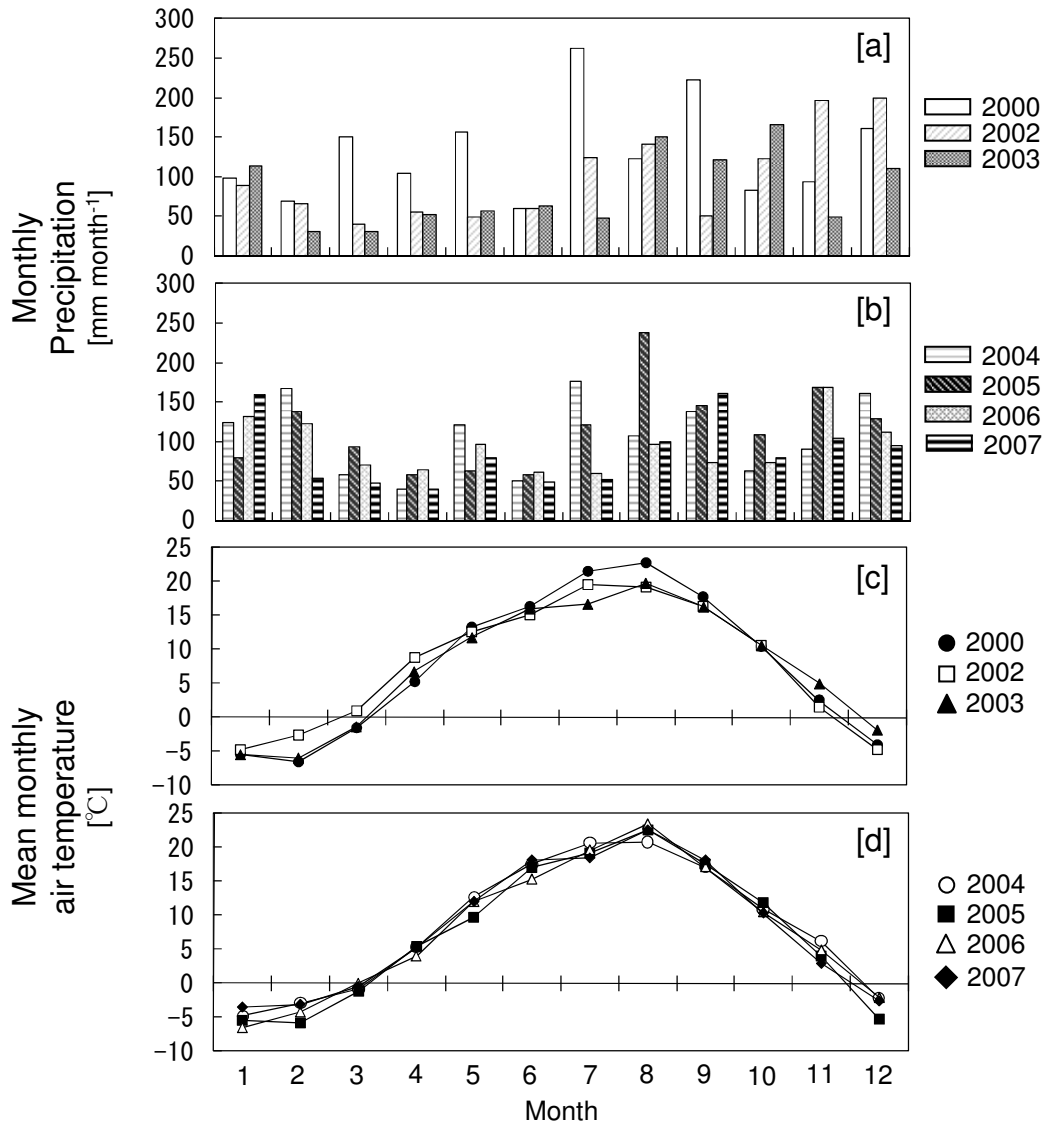


Figure 1 Seasonal variations in monthly precipitation [a, b] and mean monthly air temperature [c, d].

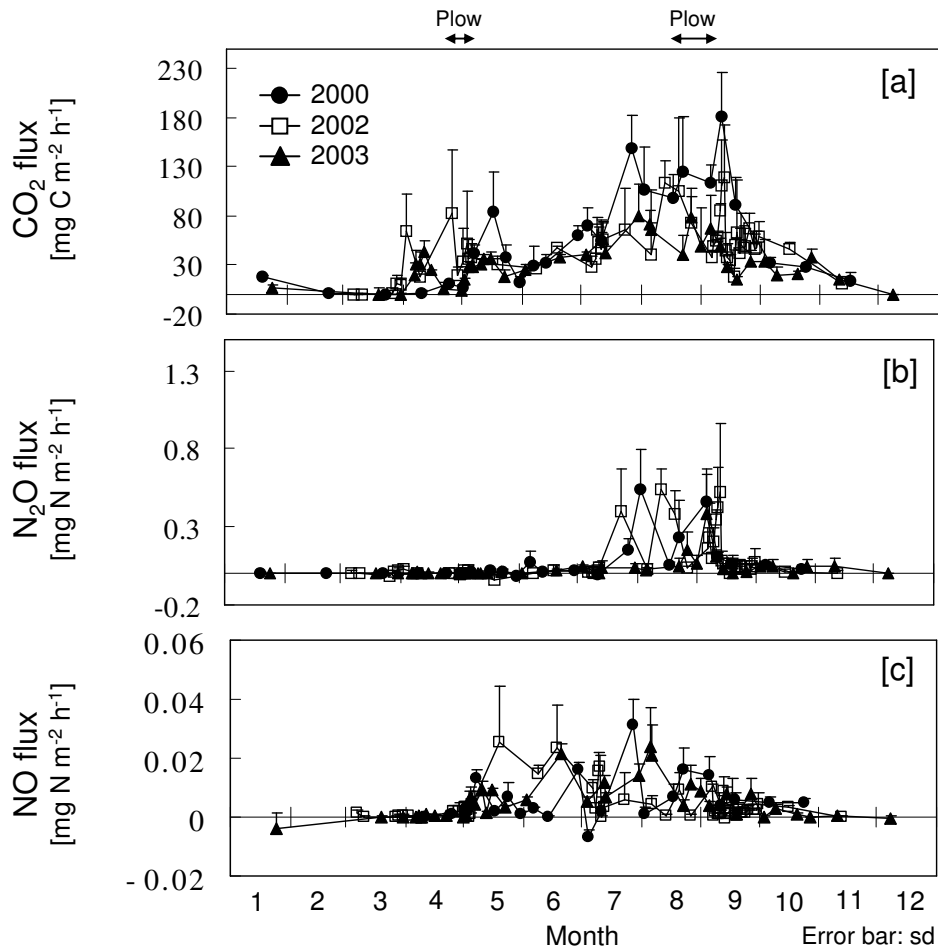


Figure 2 Seasonal variations in CO₂ [a], N₂O [b], and NO [c] fluxes in 2000, 2002, and 2003. Arrows indicate the timing of plowing.

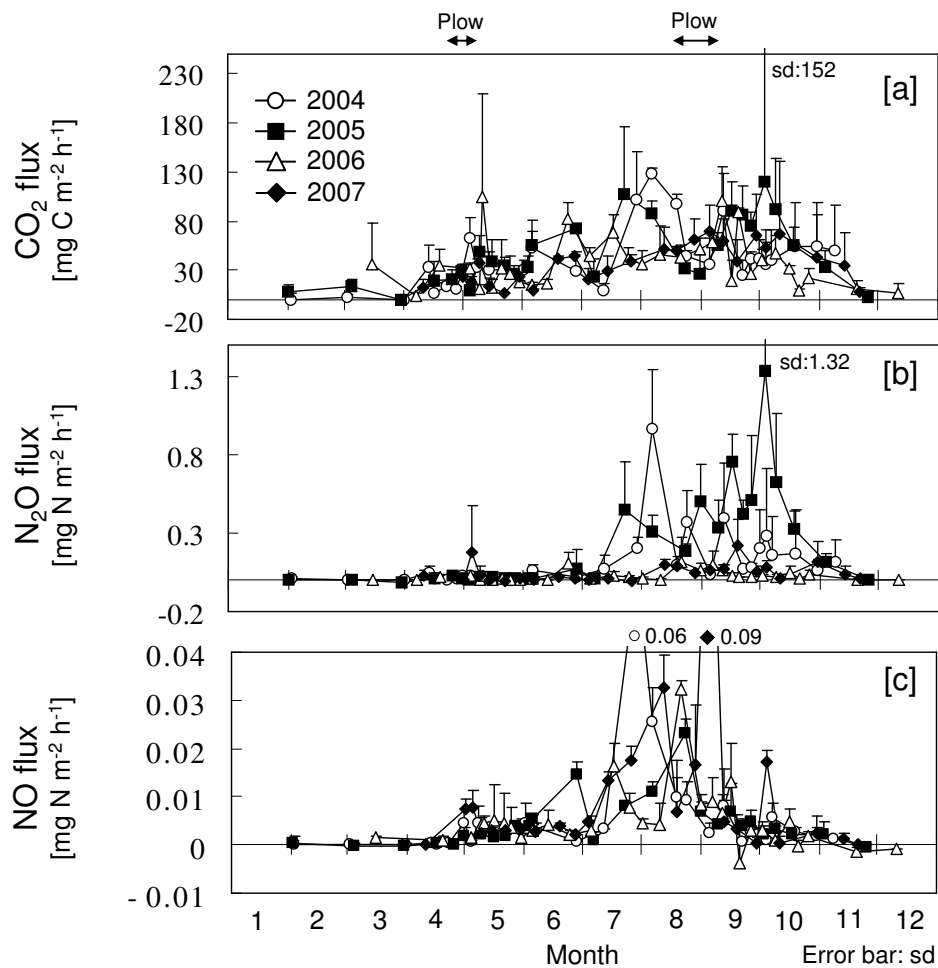


Figure 3 Seasonal variations in CO_2 [a], N_2O [b], and NO [c] fluxes from 2004 to 2007. Arrows indicate the timing of plowing.

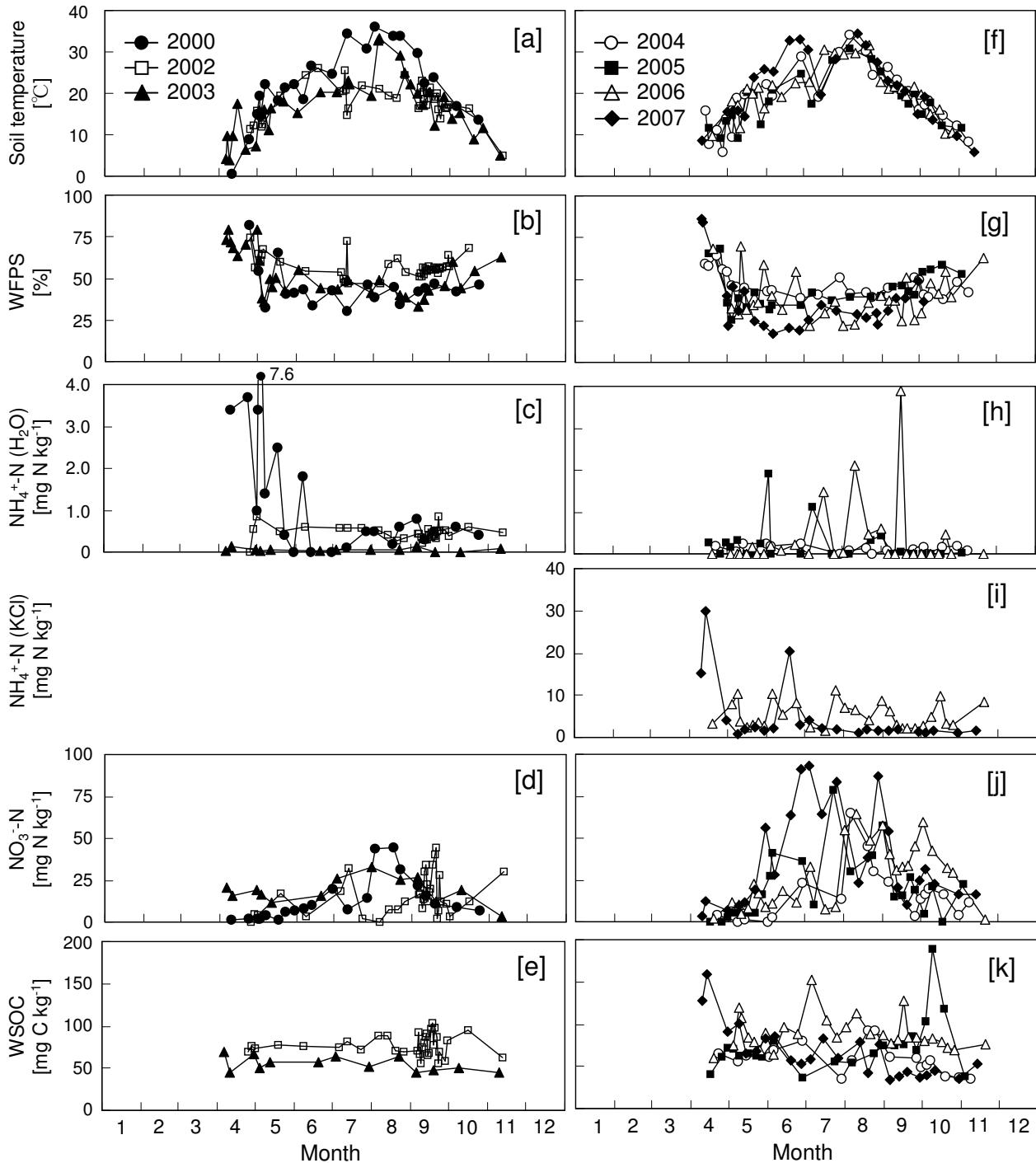


Figure 4 Seasonal variations in soil temperature [a & f], WFPS [b & g], soil $\text{NH}_4^+\text{-N}$ [c, h & j], $\text{NO}_3^-\text{-N}$ [d & i], and WSOC [e & j] concentrations.

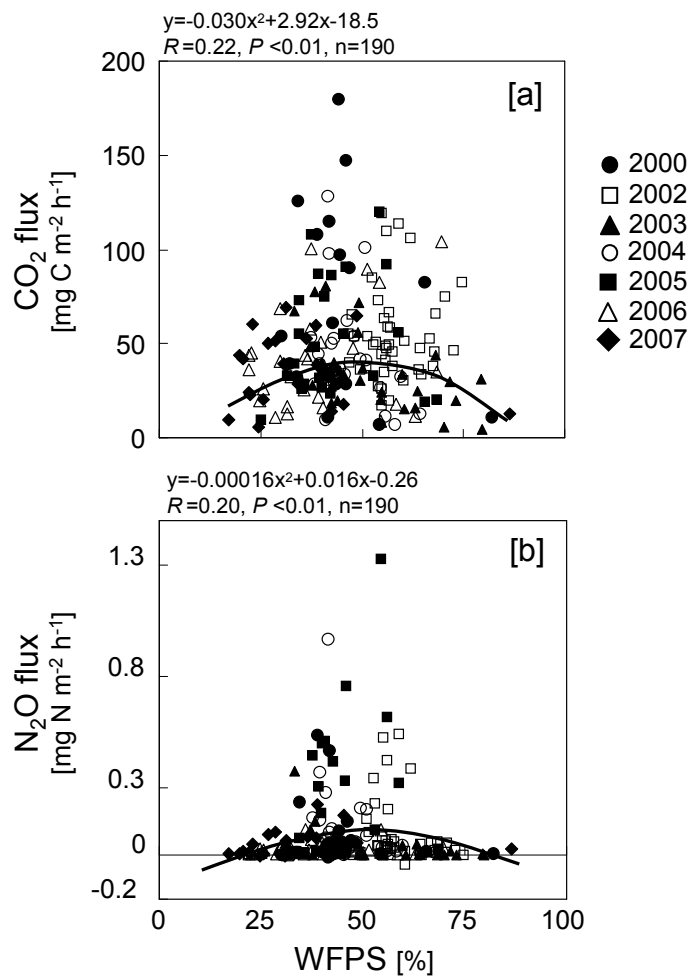


Figure 5 Relationships between CO₂ or N₂O fluxes and WFPS.

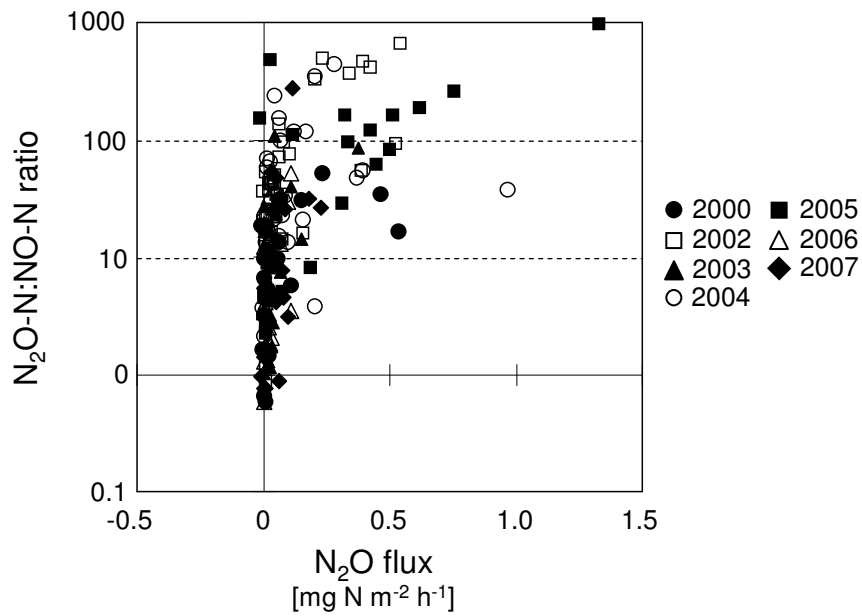


Figure 6 Relationship between N_2O flux and $N_2O-N:NO-N$ ratio.

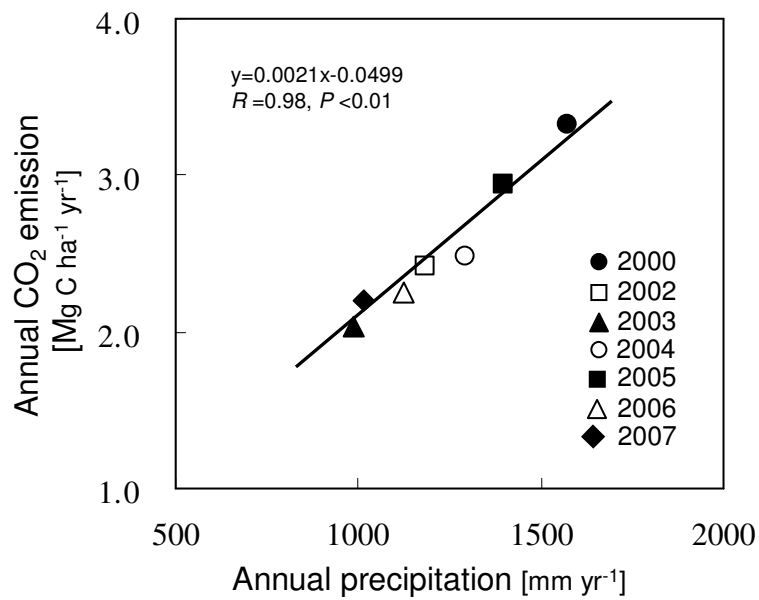


Figure 7 Relationship between CO₂ emission and annual precipitation.

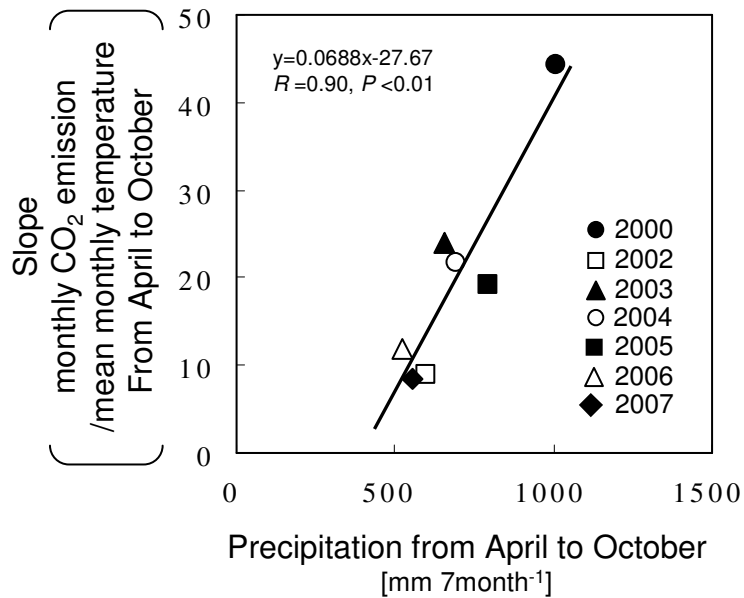


Figure 8 Relationship between slope of the monthly CO₂ emission against mean monthly temperature and precipitation from April to October.

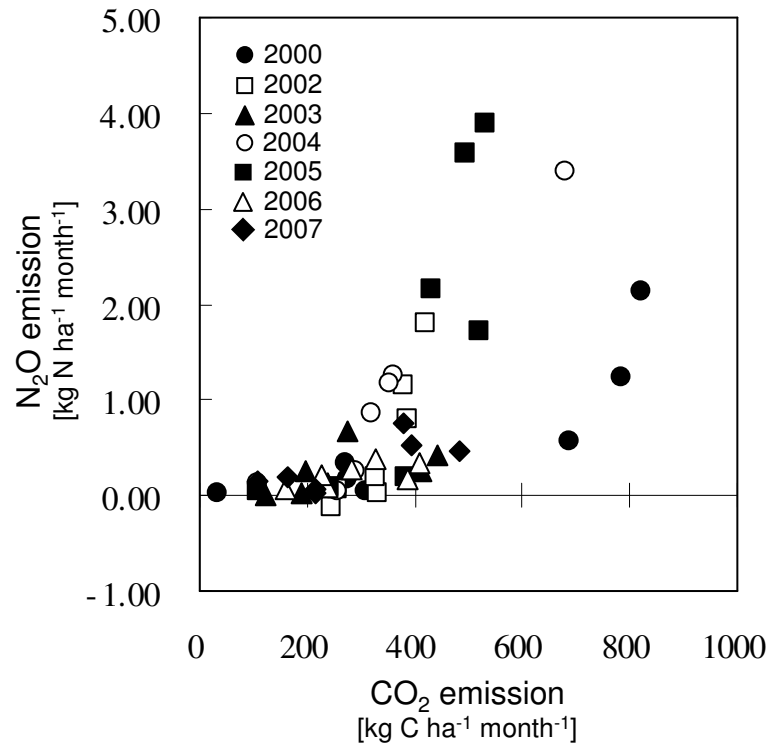


Figure 9 Relationship between monthly CO₂ emission and N₂O emission from April to October.

Table 1 Correlation coefficients of the relationships between CO₂ or N₂O fluxes and soil physical and chemical properties.

	Year	Soil temperature ^s		WFPS		NH ₄ ⁺ -N (H ₂ O)		NH ₄ ⁺ -N (KCl)		NO ₃ ⁻ -N		WSOC		CO ₂ flux	
		°C		%		mg N kg ⁻¹		mg N kg ⁻¹		mg N kg ⁻¹		mg C kg ⁻¹		mg C m ⁻² h ⁻¹	
		<i>n</i>	<i>R</i>	<i>n</i>	<i>R</i>	<i>n</i>	<i>R</i>	<i>n</i>	<i>R</i>	<i>n</i>	<i>R</i>	<i>n</i>	<i>R</i>	<i>n</i>	<i>R</i>
CO ₂ flux (mg C m ⁻² h ⁻¹)	2000	19	0.54*	19	-0.17	19	-0.25	ND	ND	19	0.56*	ND	ND	-	-
	2002	51	0.37*	43	0.09	33	-0.47**	ND	ND	33	0.08	33	0.06	-	-
	2003	34	0.68**	30	-0.51**	13	0.56*	ND	ND	13	0.66*	13	-0.27	-	-
	2004	26	0.68**	24	-0.34	18	-0.35	ND	ND	18	0.71**	18	-0.08	-	-
	2005	24	0.35	24	0.09	24	-0.37	ND	ND	24	0.33	22	0.37	-	-
	2006	27	0.31	29	0.22	29	-0.05	29	0.15	28	0.10	29	0.13	-	-
	2007	25	0.13	21	-0.11	ND	ND	21	-0.22	22	0.12	22	-0.60*	-	-
N ₂ O flux (mg N m ⁻² h ⁻¹)	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 CO₂ and N₂O emissions from unfertilized bare soil in an onion field in Mikasa

Year	Annual precipitation	Mean annual air temperature	Period day	CO ₂ emission	Unvcer- tainty	Q ₁₀	N ₂ O emission	Unvcer- tainty
	mm yr ⁻¹	°C		Mg C ha ⁻¹ yr ⁻¹	%		kg N ha ⁻¹ yr ⁻¹	%
2000	1576	7.7	302	3.32 a	14.2	2.38	4.60 bc	18.8
2002	1187	7.7	251	2.42 abc	16.9	2.03	3.94 bc	23.6
2003	986	7.4	318	2.04 c	20.7	1.80	2.01 c	74.0
2004	1294	8.4	280	2.48 bc	13.2	1.73	7.34 b	15.0
2005	1398	7.5	297	2.95 ab	21.2	1.53	12.1 a	19.0
2006	1124	7.8	271	2.25 bc	22.8	1.38	1.62 c	106
2007	1015	8.2	256	2.20 bc	58.9	1.11	2.56 c	153
Average	1226	7.81	282	2.53	24.0	1.71	4.88	58.5

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