



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
Author(s)	Toma, Yo; Kimura, Sonoko D.; Yamada, Hiroyuki; Hirose, Yuu; Fujiwara, Kazuya; Kusa, Kanako; Hatano, Ryusuke
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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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26

27 9 List of authors:
28

29
30 10 Yo Toma¹, Sonoko D. Kimura², Hiroyuki Yamada³, Yuu Hirose⁴, Kazuya
31

32 11 Fujiwara⁵, Kanako Kusa⁶, Ryusuke Hatano⁷
33

34
35 12 Institute or laboratory of origin:
36

37
38 13 ¹ Field Science Center for Northern Biosphere, Hokkaido University, Kita 11 Nishi
39

40 14 10, Kita-ku, Sapporo 060-8589, Japan
41

42
43 15 ² Graduate School of Agriculture, Department of International Environmental &
44

45 16 Agricultural Science, Tokyo University of Agriculture and Technology, Saiwaicho
46

47
48 17 3-5-8, Fuchu, 183-8509 Tokyo 183-8509, Japan
49

50
51 18 ³ Graduate School of Agriculture, Hokkaido University, Kita 9 Nishi 9, Kita-ku,
52

53
54
55
56
57
58
59
60

1
2
3
4
5
6 19 Sapporo 060-8589, Japan
7

8
9 20 ⁴ Department of Biological Science, Graduate School of Sciences, the University
10
11 21 of Tokyo, Komaba 3-8-1, Meguro, Tokyo 153-8902, Japan
12

13
14 22 ⁵ Faculty of Agriculture, Hokkaido University, Kita 9 Nishi 9, Kita-ku, Sapporo
15
16 23 060-8589, Japan
17

18
19 24 ⁶ National Agricultural Research Center, Tsukuba 305-8666, Japan
20

21
22 25 ⁷ Research Faculty of Agriculture, Hokkaido University, Kita 9 Nishi 9, Kita-ku,
23
24 26 Sapporo 060-8589, Japan
25

26
27 27

28
29 28 *Corresponding author: Yo Toma (Tel: +82-011-706-2854, E-mail:

30
31 29 toma@fsc.hokudai.ac.jp)
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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 [$\text{Mg C ha}^{-1} \text{ yr}^{-1}$] = $0.0021 \times$ 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 $\text{N}_2\text{O-N}:\text{NO-N}$ 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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19
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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23 56 was 58.5 %. Strong relationships between the monthly emissions of CO_2 and
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26 57 N_2O suggest that N_2O production by denitrification is strongly affected by SOM
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28
29 58 decomposition. Unlike the CO_2 emission, the relationship between N_2O emission
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32 59 and precipitation was not observed because of the multiple pathways of
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35 60 nitrification and denitrification for N_2O production induced by SOM
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38 61 decomposition.
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63 **Key words:** carbon dioxide, denitrification, nitrous oxide, soil organic matter
64 decomposition, temporal variation.
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6 66 **INTRODUCTION**
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8
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
12
13 69 Industrial Revolution, concentrations of these greenhouse gases in the
14
15 70 atmosphere have increased at a rate of 1.4 ppm yr⁻¹ and 0.8 ppb yr⁻¹,
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17 71 respectively (Intergovernmental Panel on Climate Change (IPCC) 2007) . The
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19 72 emission of carbon (C) that has originated due to the change in land use from
20
21 73 1980 to 1989 contributes 24% of the global annual CO₂ emission. Similarly, the
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23 74 emission of N₂O originating from agricultural fields in 1989 contributed 24% of
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25 75 the total global N₂O emission (Mosier *et al.* 1998). Recently, the annual N₂O
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27 76 emissions from fertilized cropland and grassland at a global level were estimated
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29 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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42 80 CO₂ (Hu *et al.* 2004; Koga *et al.* 2006; Koizumi *et al.* 1993; Mu *et al.* 2006, 2008;
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44 81 Shimizu *et al.* 2009). The reduction of C from soil has been estimated by a
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46 82 difference in soil C over 20 years in a 0-30 cm soil surface (IPCC 2006; Paustian
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48 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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13
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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23 90 because soil C is usually very large compared to C emission from the soil. In
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25 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
33 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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37
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
44 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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60 119 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
18
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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29
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
15
16 160 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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21 162 followed by plowing, which is carried out to incorporate the residues into the soil
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23 163 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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34 167 (three or four replications) using two types of cylindrical stainless steel chambers
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36 168 from 10:00 am to 2:00 pm. In 2000, chambers of 30 cm in diameter and 35 cm in
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38 169 height were used (Kusa *et al.* 2002, 2006), whereas chambers of 20 cm in
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40 170 diameter and 25 cm in height were used from 2002 to 2007. The cover of the
41
42 171 chamber was made of acryl and was equipped with a sample collector, a
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44 172 pressure regulating bag and a Tedlar bag (0.5 L). In 2000 and from 2002 to 2004,
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46 173 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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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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29
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
42
43 188 Oxide Analyzer (Model 265P, Kimoto Electric, Osaka, Japan), respectively. From
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45
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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6 192 $F = \rho \times (V/A) \times (\Delta c/\Delta t) \times [273/(273 + T)] \times (P/760)$
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9 193 where, F is the flux (mg N or C m⁻² h⁻¹), ρ is the gas density (ρ_{N₂O-N} = 1.259
10
11 194 × 10⁶, ρ_{CO₂-C} = 0.536 × 10⁶ and ρ_{NO-N} = 0.625 × 10⁶ mg m⁻³), V is the volume
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14 195 of the chamber (m³), A is the area of the chamber (m²), Δc/Δt is the ratio of
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17 196 change in the gas concentration inside the chamber (10⁻⁶ m³ m⁻³ h⁻¹), T is the
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19 197 air temperature inside the chamber (°C), and P is the air pressure (mm Hg; see
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21
22 198 Kusa *et al.* 2002; Toma & Hatano 2007).

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24 199 A positive value of F indicates gas emission from the soil to the atmosphere,
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27 200 while the negative value indicates gas uptake by the soil from the atmosphere.
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30 201 Considering the machinery precision, N₂O flux values within the range from -6 to
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32 202 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
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35 203 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
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38 204 annual CO₂ and N₂O emissions were calculated assuming linear changes
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41 205 between two sampling occasions.

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43 206 The value of Q₁₀, which is a relative increase of CO₂ flux for 10°C change in
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45
46 207 soil temperature, was calculated based on the Arrhenius equation and following
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49 208 equation (Hu *et al.* 2004): $k = A \times \exp(-E/RT) \rightarrow \ln k = \ln A - E/RT$
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$$Q_{10} = \frac{A \times \exp\{-E / R(T + 10)\}}{A \times \exp(-E / RT)}$$

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
10
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).
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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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6 244 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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11 246
$$\text{Uncertainty (\%)} = \{(\text{two-sided 95\% confidence interval})/2\} / \text{means} \times 100$$

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14 247 The two-sided 95% confidence interval of CO_2 and N_2O emissions were
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16
17 248 calculated by using the following equation:

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19 249
$$\text{Two-sided 95\% confidence interval} = t(\text{d.f.}, 0.05) \times \text{standard error} \times 2$$

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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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33 254 **RESULTS**

34 35 255 **Seasonal variation in CO_2 and N_2O fluxes**

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37 256 The CO_2 flux peaked from August to October (Fig. 2a, 3a), whereas the N_2O 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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45
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
50
51 261 around 50%. Soil NH_4^+ -N concentrations extracted by distilled water ranged from
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7 262 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
10
11 264 2007 did not show a distinct seasonal change (Fig. 4i), the range of
12
13
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
20
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
33 272 reached its maximum at 48.8% of WFPS (Fig. 5a). Although the N₂O flux was
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35
36 273 significantly and positively correlated with soil temperature in 2000 and 2004,
37
38
39 274 N₂O flux was significantly and positively correlated with CO₂ flux from 2000 to
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41
42 275 2006. Carbon dioxide and N₂O fluxes were significantly correlated with soil
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44
45 276 NO₃⁻-N concentration, and N₂O flux was significantly correlated with soil
46
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48 277 temperature in 2000 and 2004. However, no significant correlation was observed
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51 278 between CO₂ or N₂O fluxes and soil chemical or physical properties throughout
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54 279 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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13
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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13 301 calculated assuming linear changes between two sampling occasions. This
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15 302 indicated that the decrease in CO₂ flux during the night time, when temperature
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17 303 decreased, was not considered in calculating the annual CO₂ emission from
18
19 304 2000 to 2004. The difference in maximum and minimum air temperature from
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21 305 July to September, which was the period of high air temperature, was below
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23 306 10°C (8.8°C). In addition, the average Q₁₀ value from 2000 to 2004 was 1.99
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25 307 (Table 2). If we assume that the CO₂ flux during the maximum daily air
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27 308 temperature is 1.99 times higher than the CO₂ flux during minimum daily air
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29 309 temperature, the measured annual CO₂ emission would be 1.33 times higher
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31 310 than the annual CO₂ emission that was calculated by considering the daily
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33 311 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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47 314 the annual N₂O emission was 58.5 %. Uncertainty of yearly variation of CO₂ and
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49 315 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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29
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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45 349 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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37 364 N₂O could be the supply of C and inorganic N from SOM decomposition. Some
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40 365 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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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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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
30 433 and soil type of the study site. The mean annual temperature (13.1°C) of the
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32 434 study site reported by Koizumi *et al.* (1994) was higher than that in our study site.
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35 435 In addition, there were frequent tillage operations because of a double-cropping
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37 436 system. High temperature and frequency of plowing may have increased SOM
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39 437 decomposition in the study reported by Koizumi *et al.* (1994). On the other hand,
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41 438 the study site of Koga *et al.* (2006) was located in similar latitude (42°53 'N) as
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43 439 this study. Therefore, difference in SOM decomposition between our study and
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45 440 the study conducted by Koga *et al.* (2006) might be due to the differences in crop
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48 441 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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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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39 509 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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42 510 ha⁻¹ yr⁻¹ (Zou *et al.* 2005). The difference in annual N₂O emission might be due to
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45 511 soil physical and chemical properties. Nitrogen content in soil may affect N₂O
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48 512 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. Klemmedtsson *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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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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24
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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6 550 nitrification processes, oxygen is required for oxidation of organic C or NH₄.
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9 551 However, an anaerobic condition is required for denitrification because NO₃ is
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11 552 used as an electron capture (Bouwman and Boumans 2001). Since N₂O 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 N₂O 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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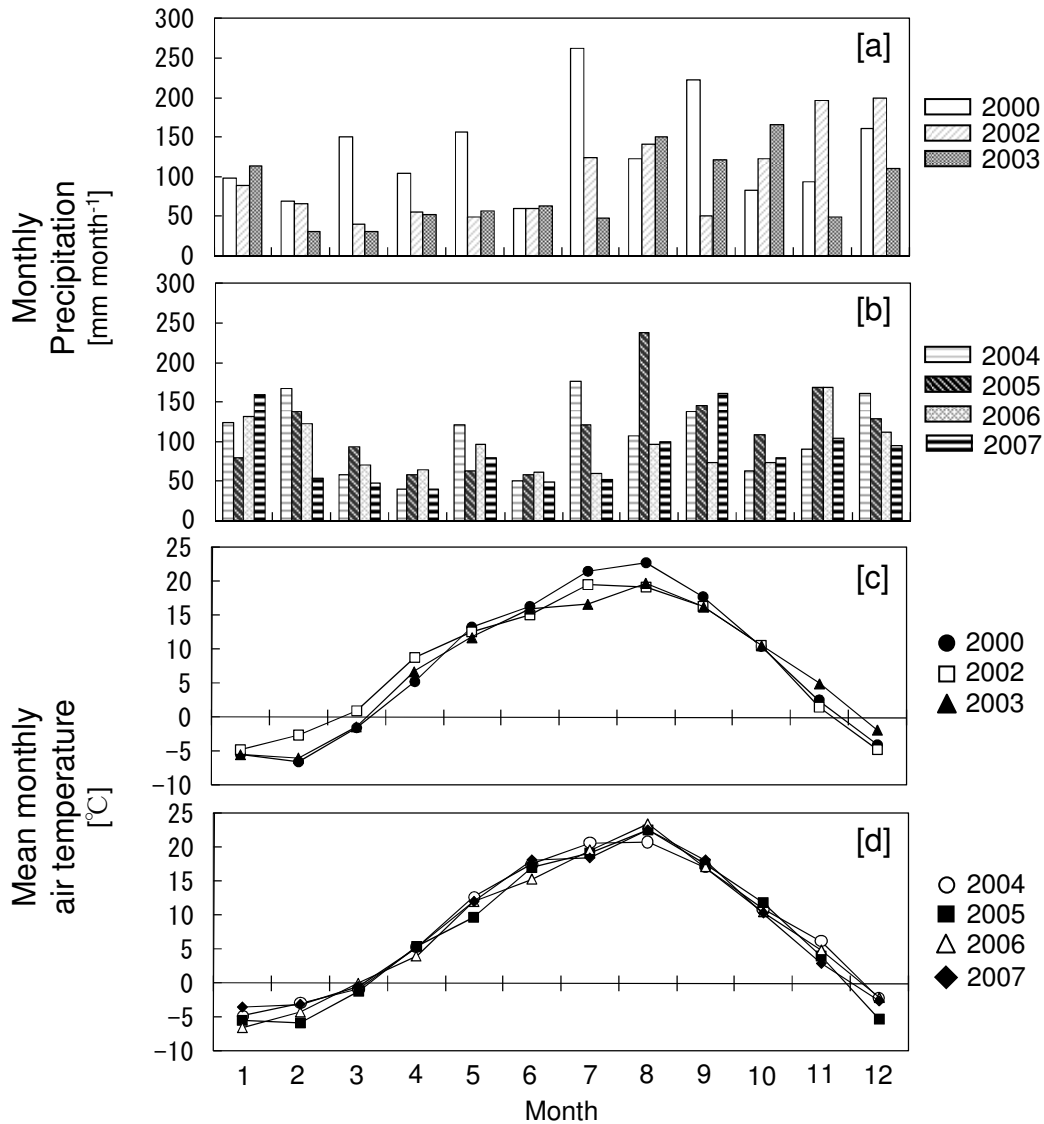


Figure 1 Seasonal variations in monthly precipitation [a, b] and mean monthly air temperature [c, d].

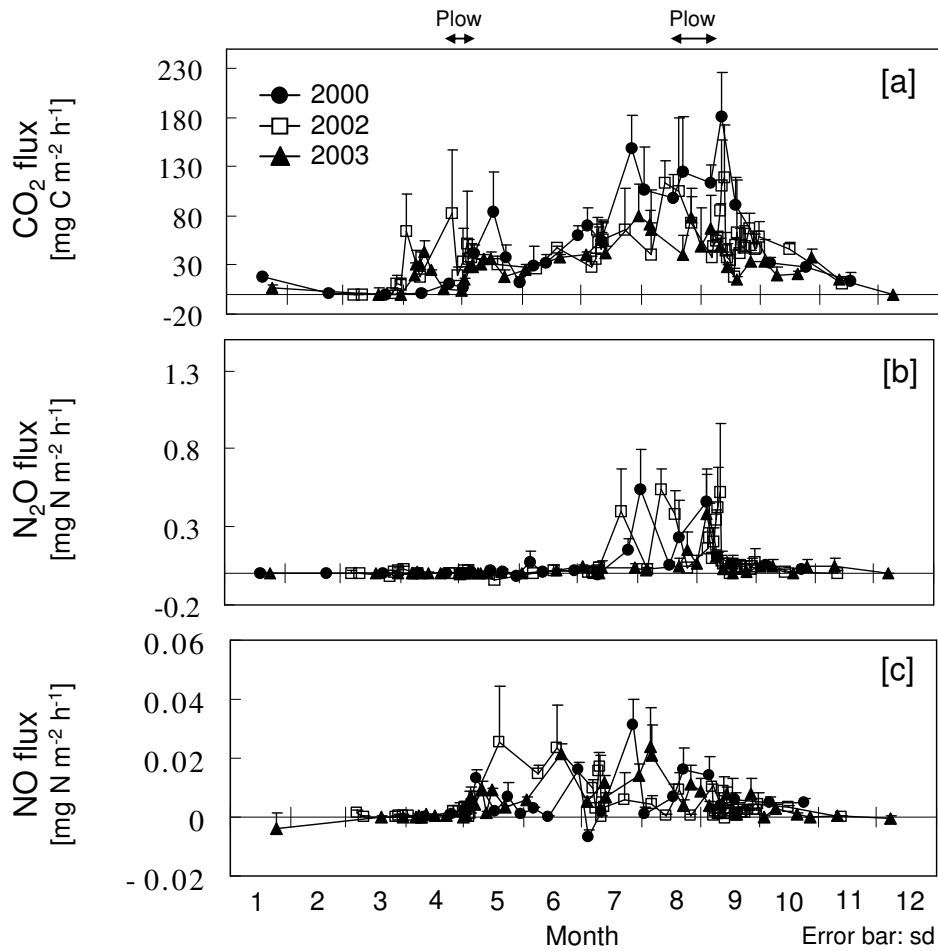


Figure 2 Seasonal variations in CO₂ [a], N₂O [b], and NO [c] fluxes in 2000, 2002, and 2003. Arrows indicate the timing of plowing.

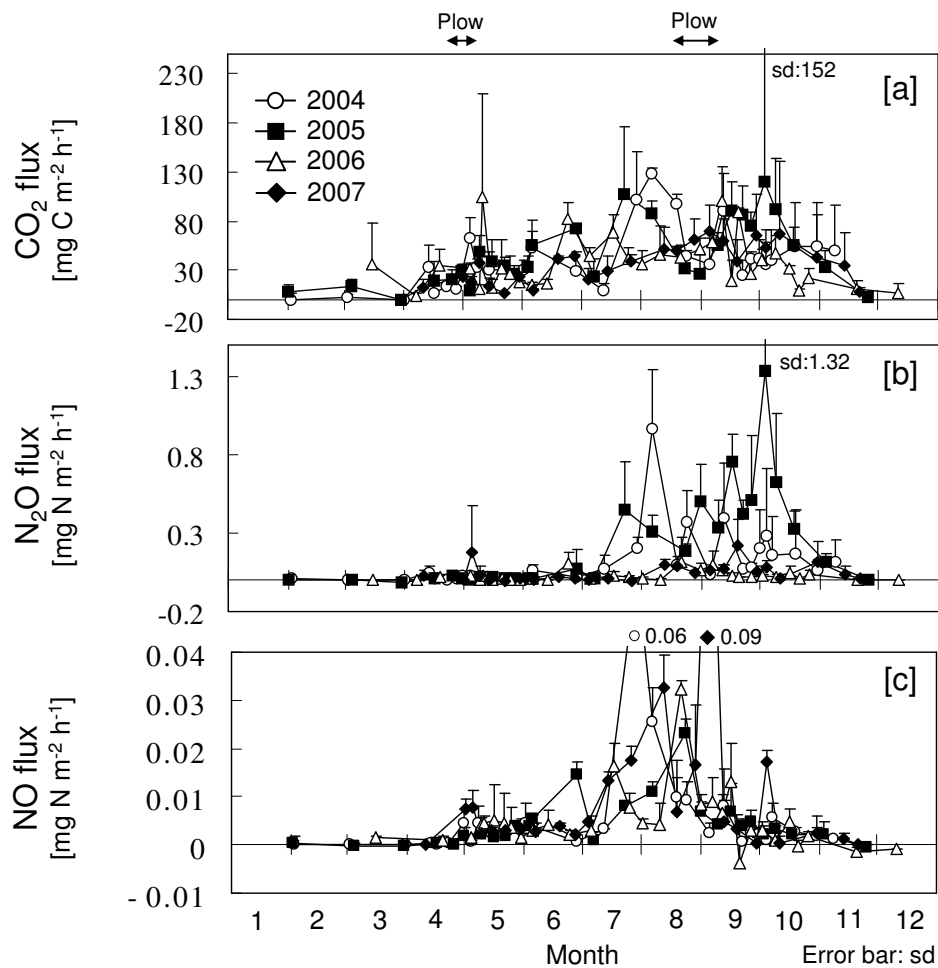


Figure 3 Seasonal variations in CO₂ [a], N₂O [b], and NO [c] fluxes from 2004 to 2007. Arrows indicate the timing of plowing.

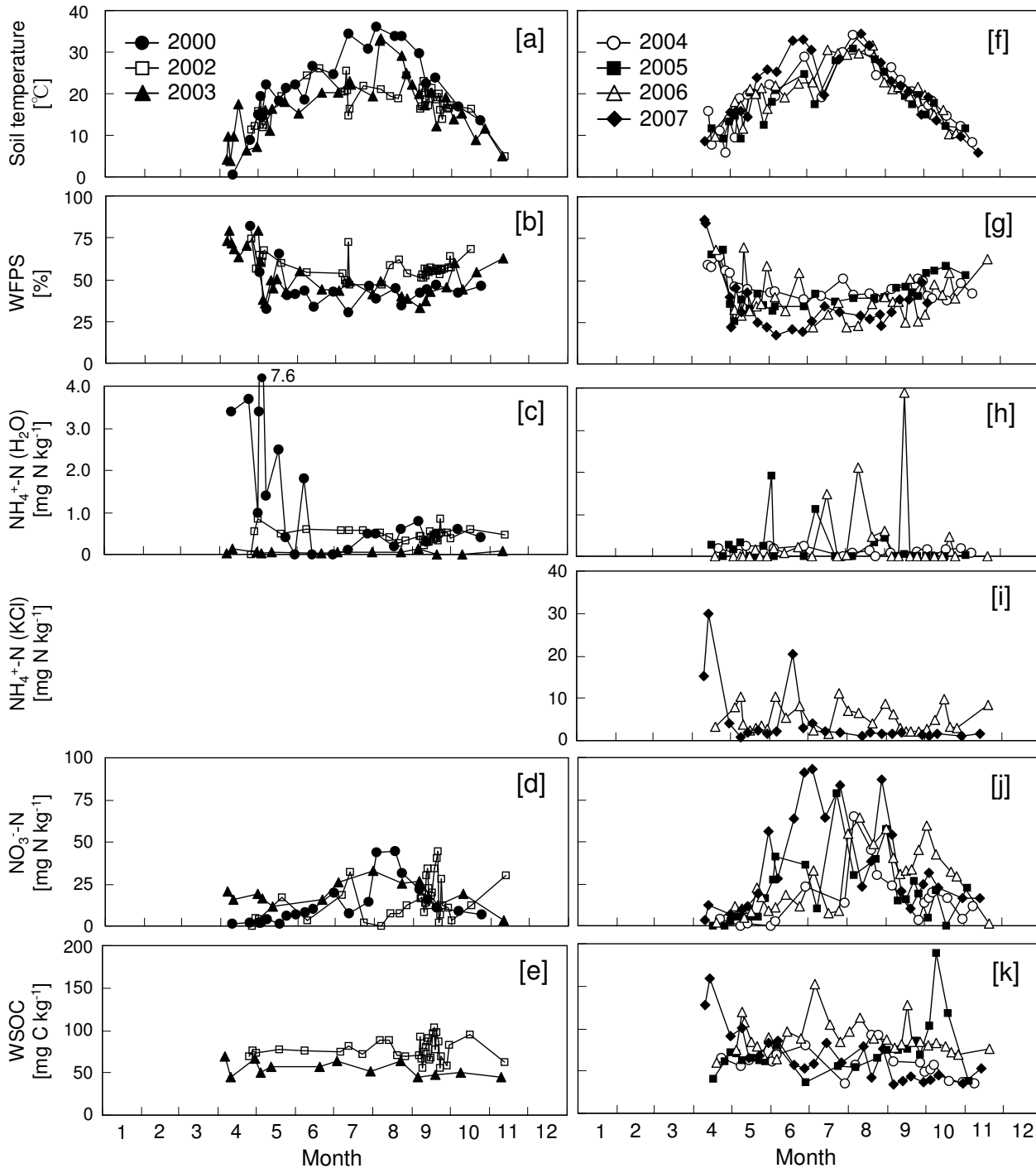


Figure 4 Seasonal variations in soil temperature [a & f], WFPS [b & g], soil $\text{NH}_4^+\text{-N}$ [c, h & j], $\text{NO}_3^-\text{-N}$ [d & i], and WSOC [e & j] concentrations.

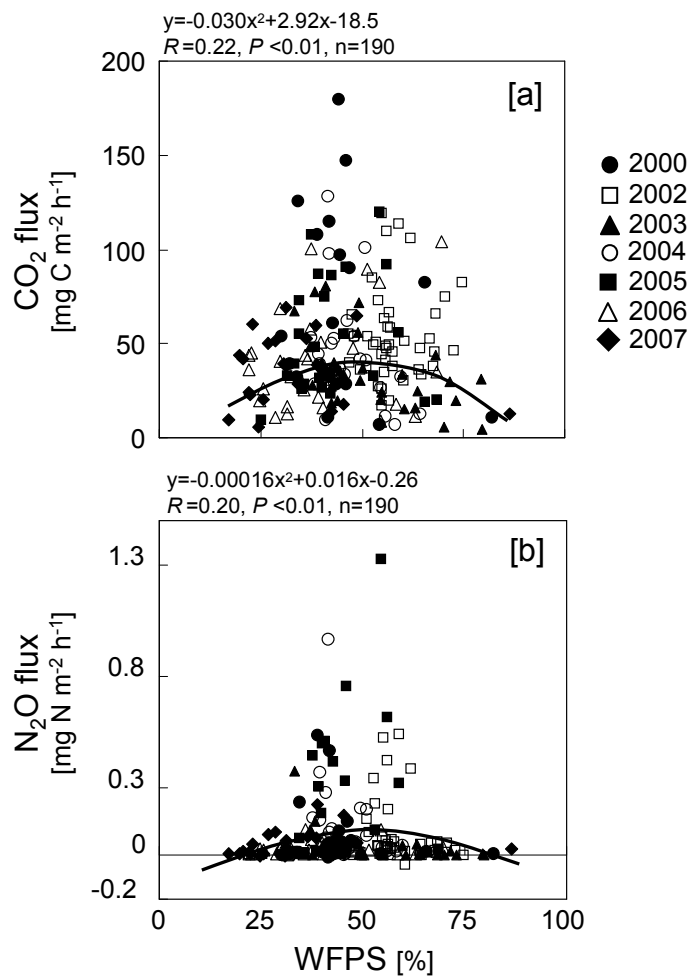


Figure 5 Relationships between CO₂ or N₂O fluxes and WFPS.

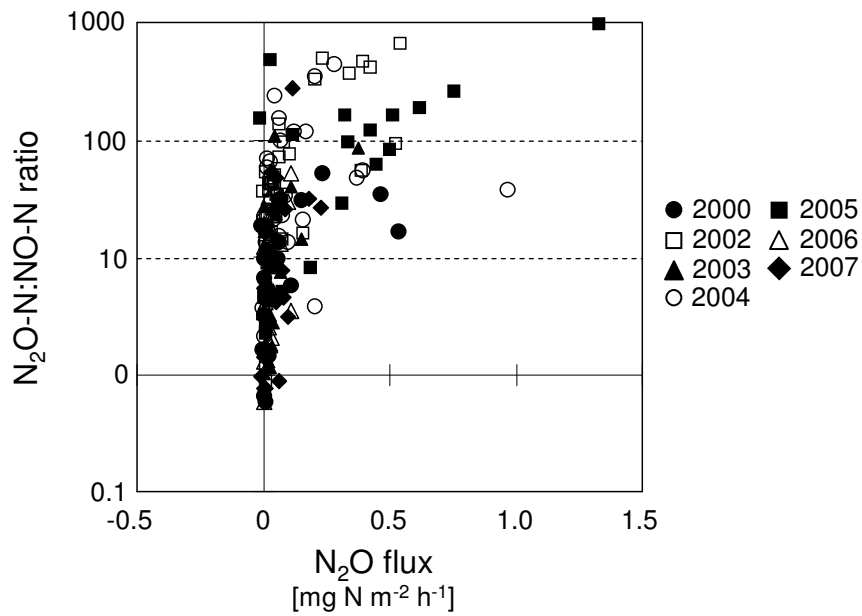


Figure 6 Relationship between N_2O flux and $\text{N}_2\text{O-N:NO-N}$ ratio.

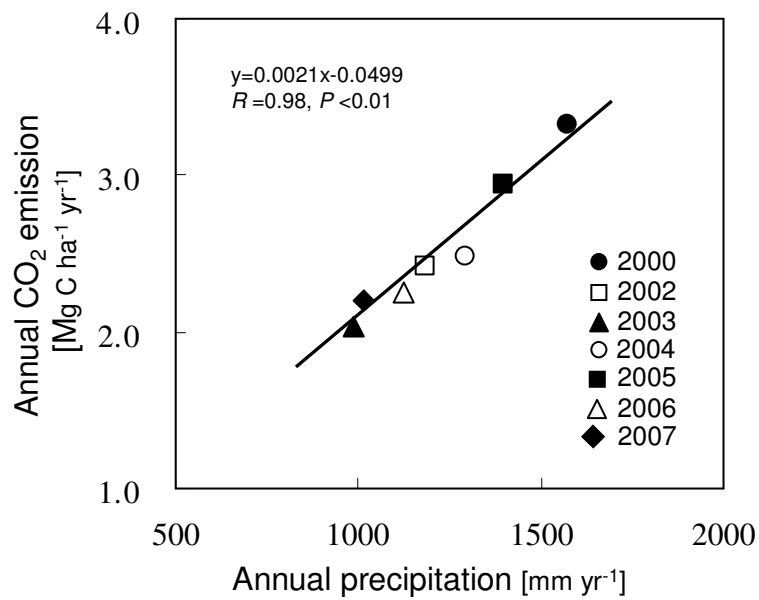


Figure 7 Relationship between CO₂ emission and annual precipitation.

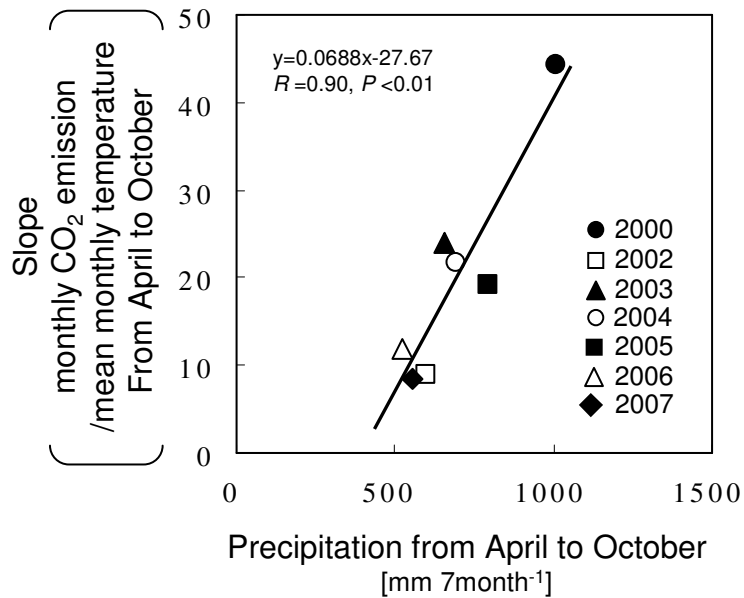


Figure 8 Relationship between slope of the monthly CO₂ emission against mean monthly temperature and precipitation from April to October.

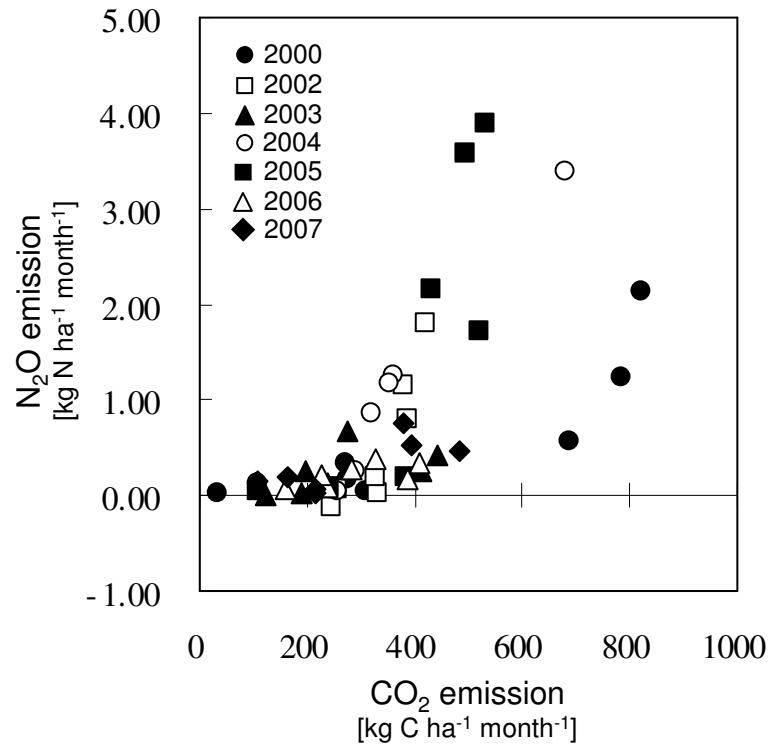


Figure 9 Relationship between monthly CO₂ emission and N₂O emission from April to October.

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 ($P < 0.05$)