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TITLE:
N_2O and NO fluxes from cornfield, grassland, pasture, and forest in a watershed in Southern Hokkaido, Japan

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ABSTRACT

To develop an advanced method for estimating nitrous oxide (N_2O) emission from an agricultural watershed, we used a closed-chamber technique to measure seasonal N_2O and nitric oxide (NO) fluxes in cornfields, grassland, pastures, and forests at the Shizunai Experimental Livestock Farm (467 ha) in southern Hokkaido, Japan. From 2000 to 2004, N_2O and NO fluxes ranged from -137 to 8920 $\mu g\ N\ m^{-2}\ h^{-1}$ and from -12.1 to 185 $\mu g\ N\ m^{-2}\ h^{-1}$, respectively. Most N_2O/NO ratios calculated on the basis of these N_2O and NO fluxes ranged between 1 and 100, and the log-normal N_2O/NO ratio was positively correlated with the log-normal N_2O fluxes ($r^2 = 0.346$, $P < 0.01$). These high N_2O fluxes therefore resulted from increased denitrification activity. Annual N_2O emission rates ranged from -1.0 to 81 kg N $ha^{-1}\ y^{-1}$ (average = 6.6 kg N ha^{-1}). Since these emission values varied greatly and included extremely high values, we divided them into two groups: normal values (i.e., lower than the overall average) and high values (i.e., higher than average). The normal data were significantly positively correlated with N input ($r^2 = 0.61$, $P < 0.01$) and the "higher" data from ungrazed fields were significantly positively correlated with N surplus ($r^2 = 0.96$, $P < 0.05$). The calculated probability that a high N_2O flux would occur was weakly and positively correlated with precipitation from May to August. This probability can be used to represent annual variation in N_2O emission rates and reduce the uncertainty of N_2O estimation.

Key words: nitrous oxide, nitric oxide, outliers, uncertainty, watershed

INTRODUCTION

Nitrous oxide (N_2O) is a major greenhouse gas. Over a 100-year time horizon, it has 296 times the global warming potential of CO_2 (IPCC 2001), and it is also responsible for the destruction of stratospheric ozone (Crutzen 1970). The N_2O concentration in the atmosphere rose from a pre-industrial level of 270 ppb to 314 ppb in 1998 (IPCC 2001). Soils are a major source of N_2O, and the global N_2O emission rate from soils has been estimated at 10.2 Tg N y^{-1}, equivalent to 58% of the total N_2O emission rate of 17.7 Tg N y^{-1} (IPCC 2001). However, estimates of the global N_2O emission rate from soils contain large uncertainties.

In soils, N_2O is produced mainly by the microbial processes of nitrification and denitrification. These biological processes are affected by soil environmental factors such as moisture conditions, oxygen status, soil temperature, N availability, organic matter content, and pH (Sahrawat & Keeney 1986; Mosier 1998; Wrage *et al.* 2001). It is well known that these soil environmental factors have a large variability in space and time (e.g., Parkin 1993); this makes it difficult to estimate soil N_2O emission rates on a large scale (Mosier 1998).

Countries listed in Annex I of the United Nations Framework Convention on Climate Change (UNFCCC) are required to submit national greenhouse gas inventories to the UNFCCC secretariat. To meet this requirement, it is necessary to estimate N_2O emission rates from soils by choosing the best method that has currently been proposed by IPCC guideline, even if the estimates by this method contain considerable uncertainty. Sozanska *et al.* (2002) estimated N_2O emission rates from soils in Great Britain by using published N_2O data and a GIS framework. They developed a regression model based on N_2O data, N input, water-filled pore space (WFPS), soil temperature, and land-use type. They reported that N input was the main explanatory variable in their regression model, and that soil moisture conditions also strongly affected N_2O emissions. Kaiser and Ruser (2000) reported that N_2O emission was explained better by the N surplus than by the N input. They summarized 99 long-term field studies on N_2O emission rates in Germany and found no significant relationship between annual N_2O emission and the rate of application of N fertilizers. However, they reported that the N surplus over a 4-year period was a suitable predictor of N_2O. In Japan, Akiyama *et al.* (2006) compiled N_2O emission rates from Japanese agricultural fields, and calculated emission factors. They classified these measured data into those measured in upland, tea, and paddy fields. The measured data for upland fields were divided into data collected from well-drained and poorly drained soils.

The abovementioned studies reported the estimated N_2O emission rates for a wide area by using measured N_2O emission rates, N inputs or surpluses, and data on soil moisture conditions. However, the estimates derived by the methods proposed in these studies contain considerable uncertainty due to the wide variations in the observed N_2O emission rates. In addition, some of the proposed models excluded high N_2O emission rates (i.e., “outliers”), despite the fact that high N_2O emission rates strongly influence the overall N_2O estimates and increase the range of the estimated values. This is a significant problem, because high N_2O natural emission rates are occasionally observed under certain combinations of environmental conditions (Sahrawat & Keeney 1986), indicating that the outliers may represent valid data rather than measurement or modeling errors. In addition, these models cannot represent the annual variation in N_2O emission rates. For a proper representation, continuous measurements should have been taken over a period of several years to detect inter-annual variation. Therefore, there is clearly room to develop a new method that can more accurately estimate N_2O emission rates throughout the year.

As a first step in developing such a method, we measured N_2O fluxes over a 5-year period in cornfields, grassland, pastures, and forests within an agricultural watershed. To develop a new method for N_2O estimation, we evaluated N_2O emission rates by using N budget data, soil moisture conditions, and meteorological data. Here we report some of the characteristics of our model for estimating N_2O emission rates.

MATERIALS AND METHODS

Study area

The study was carried out at the Shizunai Experimental Livestock Farm (Fig. 1) of the Field Science Center for Northern Biosphere, Hokkaido University, Japan (42˚25'9"N, 142˚29'1"E). In addition to being an experimental station, the farm is also a working production facility where young animals are reared to maturity (e.g., for beef production and species preservation of Hokkaido native horses) and crops (primarily corn and grass) are grown to support the animals. The altitude of the farm ranged from 40 to 360 m asl, with the southern and southeastern parts being lower than the other parts of the farm. The farm is located in the watershed of the Kepau River, which flows through the farm from an upstream forested area. N cycling at the livestock farm has been well investigated (Hayakawa *et al*. 2004; Hatano *et al*. 2005). The annual mean temperature at the study site is 7.9 °C, the monthly minimum temperature is –8.1 °C (February), and the monthly maximum temperature is 23.6 °C (August). The monthly mean temperature from July to September (2000 to 2004) was higher than that during the other months (Fig. 2). The annual mean precipitation is 1365 mm. From 2000 to 2004, the annual variation in monthly precipitation was high, and the monthly precipitation was higher from May to September (except for June) than during the remaining months (Fig. 2). The major soil types are a Vitric Andosol and a Histosol (FAO 1988) (Table 1).

The total area of the livestock farm is 458 ha. The total area of each land-use type during the study period was 10.3 ha for cornfields (two sites), 37.0 ha for grassland (10 sites), 102 ha for pastures (36 sites), and 308 ha for forest (12 sites). The area of each land-use type was essentially constant between 2000 and 2004, except for the cornfields (which decreased to 7 ha) and the grassland (which increased to 39.6 ha) in 2002. The dominant vegetation for each land-use type were *Zea mays* L. in the cornfields; *Phleum pratense* L., *Phalaris arundinacea* L., and *Trifolium repens* L. in the grassland; *Dactylis glomerata* L. and *T. repens* in the pastures; and deciduous broad-leaved trees (dominated by *Ulmus davidiana* Planch. var. *japonica* (Rehd.) Nak., *Quercus cuspidata* Blume, and *Acer mono* Maxim. var. *marmoratum* (Nichols.) Hara f. *dissectum* (Wesmael) Rehd.) in the forest and perennial vegetation on the forest floor. The livestock farm maintains about 150 head of beef cattle, 70 native Hokkaido horses, and 10 racehorses. The grazing season for beef cattle and racehorses is from May to October but is year-round for native horses. Inorganic fertilizers are usually applied to the cornfields in mid-May, to the grasslands in May and July, and to the pastures in July or August. Manure and slurry are usually applied to the cornfields and the grassland in early spring (February to April) and autumn (mainly October to November), and to the grassland in August after harvesting. The corn is harvested mainly in October, and the grass is harvested twice a year (July to August and August to October).

Estimating N budgets

Hayakawa *et al*. (2004) estimated annual N budgets for each site at the farm for the 5-year period from 2000 to 2004. The input and output of N were calculated mainly from the daily management records, in which management information of the livestock and of the fields were written down. When using N budgets data for this paper, we considered the quantity, variety, and timing of the application of inorganic fertilizer, manure, and slurry, the feed supply, and the grazing and harvesting of grasses. Ammonia volatilization, denitrification, N fixation, and N deposition were not accounted for in the current estimation process, but will be considered in future revisions of our approach.

Other N inputs included inorganic fertilizer, manure, slurry, and livestock excreta during grazing. The application of inorganic fertilizer was estimated by multiplying the amount that was applied by the N content (the certified value provided by the manufacturer). The total applied manure and slurry N was calculated by multiplying the number of trucks (3 t fresh matter truck^{-1}) by the N content per truck (at rates of 0.57 and 0.36 % for fresh matter of manure and slurry, respectively; Matsumoto *et al*. 2002). Livestock excreta produced during grazing were calculated by subtracting the body increment N in the livestock and milk N from the N supplied by grazing. The method of calculation for livestock excreta was described in Hayakawa *et al*. (2004) in detail.

The N output included the yield N in the grass and corn as well as the N in grazed grass. The yield N in the grass and corn was calculated by multiplying the dry weight of the rolls of hay or harvested corn in each field (both obtained from the daily management records) by the corresponding N content value (MAFF 1995). The N content in grazed grass was calculated by the following equation:

$$\text{Grazing nitrogen (kg N d}^{-1}\text{)} = [ME_{\text{grazing}} / ME_{\text{pasture}}] \times N_{\text{pasture}} \qquad (1)$$

$ME_{grazing}$ is the metabolic energy of livestock during the grazing period (MJ d^{-1}); it can be estimated by the livestock energy demand (MAFF 1995; Equine Research Institute 1998) using average live weight values (W_{ave}, kg) during the period and the daily gain in weight (DG, kg d^{-1}) of each animal. ME_{pasture} is the metabolic energy of pasture (MJ kg^{-1} DM; MAFF 1995). N_{pasture} is the N content in pasture vegetation (% DM; Hata 2000; MAFF 1995).

The N surplus for each site was estimated by subtracting the total outputs from the total inputs.

Nitrogen input and output values for the sites where N_2O fluxes were measured are shown in Table 2. (The area of F_1 was left blank in this table because this site was outside the experimental site, and was adjacent to the livestock farm.) Nitrogen input differed significantly ($P < 0.05$) among land-use types; it was highest for the cornfields, followed by the grassland and pasture, and was lowest for the forests. There was no measurable N input to the forested sites except F_2, where native horses grazed from 2001 to 2003. Sites C_2, G_1, G_2, and G_3 were also grazed by native horses in autumn, so there was an input of excreta N to these sites (Table 2). Nitrogen output was greater from the cornfields and grassland than from the forest ($P < 0.05$). There were no N outputs from the CG site (grassland created from a cornfield in 2003) in 2003, because the field was seeded in August. Fertilization was carried out in May and June, and harvesting took place in June and August in 2004. Most of the sites showed surplus N (Table 2). However, sites G_3 and F_2 showed zero or negative values throughout the research period. This resulted from low manure N inputs at site G_3 and from feeding by horses at site F_2.

Measuring seasonal N_2O and NO fluxes

Field measurements were conducted throughout the 2000–2004 period, except during the snowfall period (January to April). Seasonal N_2O and NO fluxes were measured at 12 sites, including two cornfields, four grassland sites, three pastures, and three forested sites (shown in Fig. 1 and table 1). Gas flux measurements were carried out each year at one to three sites for each land-use type. Soil properties of the measured sites are given in Table 1. The soil's total carbon content was higher at the forested or Histosol sites than in the other land-use or soil types. The soil bulk density was higher in the pastures than in the other land-use types.

N_2O and NO fluxes from the soil surface were measured more than once a month by the closed-chamber technique of Rolston (1986). The frequency of measurement in the cornfields and grassland was increased to twice a week after fertilization to capture any short-term fertilization effects. Each flux measurement was replicated three to six locations per site. The methods of measuring and calculating gas fluxes were described in detail in our previous paper (Katayanagi & Hatano 2005). We used the measured N_2O and NO fluxes to calculate the N_2O/NO ratio whenever both N_2O and NO were positive values.

Calculation of annual N_2O and NO emission rates

The annual N_2O and NO emission rates were calculated by linear interpolation of the average N_2O and NO fluxes (μg N m^{-2} h^{-1}) between the measurements and summing the results over the total time period. We defined the cumulative N_2O and NO emission rates from May to December as the annual rates. N_2O and NO fluxes were measured at two positions at site G_1 (G_{1t} [top] and G_{1b} [bottom] , at the upper and lower parts of the site, respectively) in 2002, three positions at G_2 (the G_{2s} [south], G_{2c} [center], and G_{2n} [north] parts of the field) in 2004, and four positions at F_3 (F_{3h} [boundary between G_1 and F_3; edge of the forest], F_{3d} [depression near the Kepau River], F_{3r} [the riverside], and F_{3f} [inside the forest]) from 2002 to 2004. When annual N_2O and NO emission rates were calculated, these positions were considered separately.

Measuring and calculating soil properties

Volumetric water content (*VWC*, m^3 m^{-3}) of the soils to a depth of 10 cm was measured (four replications) by means of time-domain reflectometry (TDR; TRIME-FM, probe-P2). Water-filled pore space (WFPS, %) of the soils was calculated from the measured VWC by the following equation:

$$WFPS\ (\%) = [measured\ VWC / porosity] \times 100 \qquad (2)$$

where the soil porosity (m^3 m^{-3}) at each site is shown in Table 1. The soil porosity were measured by a three phase meter (Daiki Rika Co. Ltd., DIK-1110).

Statistical analyses

Significant differences between the mean values of measured N_2O emission rates from the various land-use types were evaluated by means of Tukey tests ($P < 0.05$). The relationship between N_2O fluxes and WFPS were estimated by the boundary-line approach (Webb 1972). In this method, a regression curve was described by connecting the data points at the outer margin of the data. This approach is based on the hypothesis that the outer margin of the data depicts the limits of the functional dependency between the two factors (in this case, N_2O flux and WFPS). Schmidt *et al.* (2000) describe the boundary-line approach in detail.

RESULTS

Temporal variability in soil temperature and WFPS

Soil temperatures at 5- and 10-cm depths increased from March to August and decreased from August to December, following a similar pattern to that of air temperature (Fig. 3). Soil temperature at 5 cm was higher than that at 10 cm for all measured sites, especially in June and July.

WFPS of the soils to a depth of 10 cm showed temporal variability and depended on precipitation and temperature (Fig. 4). It increased after rainfall and decreased after consecutive sunny, warm days. The soils were generally saturated (100% WFPS) just after a heavy rainfall. The annual WFPS to a depth of 10 cm averaged (±SD) 61%±9% in the cornfields, 78%±8% in the grasslands, 80%±15% in the pastures, and 59%±11% in the forests.

Temporal variability in N_2O and NO fluxes

The temporal variability in N_2O flux was large (Fig. 5). N_2O fluxes ranged from -137 to 8920 μg N m^{-2} h^{-1}. High N_2O fluxes were observed during various periods, and their magnitude varied greatly among the years. For example, the highest N_2O emission rates from the cornfield that was subsequently converted into a grassland (CG) and the grassland (G_{1t}) were 531 and 204 μg N ha^{-1} y^{-1}, respectively, in 2000, versus 8920 and 4713 μg N ha^{-1} y^{-1} in 2002. High N_2O fluxes from cornfields, grassland, and pastures were observed mainly during the growing period, from May to October, and the fluxes were especially high from July to August (Fig. 5). During this period, fertilizers were applied and large amounts of precipitation occurred (Fig. 5). N_2O fluxes from the forests were lower (averaging near zero, and with maximum values less than about 150 μg N ha^{-1} y^{-1}) than those from the other land-use types.

Similarly to the trend for N_2O, NO fluxes also showed a large temporal variability. NO fluxes ranged from -12.1 to 185 μg N m^{-2} h^{-1}. However, the magnitude of the variation was smaller than that of the N_2O fluxes (Fig. 6). Except at the forest sites (where values remained near zero), large NO fluxes were observed mainly after fertilization, and especially after the basal fertilization in May. NO emission rates from forest sites were lower than those from the other land-use types.

N_2O/NO ratio

The temporal variability in the N_2O/NO ratio did not show a clear pattern at any of the sites (Fig. 7). However, most of the values were distributed from 1 to 100, and a positive correlation ($r^2 = 0.35$, $P < 0.01$) was found between the log-normal measured N_2O fluxes and the log-normal N_2O/NO ratios (Fig. 8).

Relationship between N_2O flux and WFPS

A relationship between the N_2O fluxes and WFPS of the soils to a depth of 10 cm was observed when the boundary-line approach was applied to the data (Fig. 9). The WFPS values at maximum N_2O flux were 58% for the cornfield, 56% for the forests, 74% for the grassland, 72% for the pasture, and 67% for all land-use types combined.

Annual N_2O and NO emission rates

Annual N_2O emission rates, calculated from the measured N_2O fluxes, ranged from -1.0 to 80.8 kg N ha^{-1} y^{-1} (Table 3). The average of these values over the full study period was 6.6 kg N ha^{-1} y^{-1} ($n = 46$) and the background N_2O emission rate was 0.30 kg N ha^{-1} y^{-1} ($n = 15$). Here, the background N_2O emission rate was defined as the average for the forest sites that received no artificial N input (i.e., excluding site F_2 after 2000) during the research period. High annual N_2O emission rates were strongly influenced by high N_2O fluxes. A significantly positive correlation was found between the highest N_2O fluxes during a year and the annual N_2O emission rates from each site ($r^2 = 0.98$, $P < 0.01$; Fig. 10).

Annual N_2O emission rates from each land-use type ranged from 4.9 to 80.8 kg N ha^{-1} y^{-1} for the cornfields, from 1.1 to 42.8 kg N ha^{-1} y^{-1} for the grassland, from 1.7 to 20.3 kg N ha^{-1} y^{-1} for the pastures, and from -1.0 to 1.7 kg N ha^{-1} y^{-1} for the forests (Table 3). A comparison of annual N_2O emission rates among the land-use types showed that significantly higher annual N_2O emission rates ($P < 0.01$) were observed from the cornfields than from the other land-use types; the latter did not differ significantly each other (Table 3).

N_2O fluxes were measured at two positions at site G_1 in 2002, three positions at site G_2 in 2004, and four positions at site F_3 from 2002 to 2004. Although each site was managed uniformly, we observed greatly different N_2O emission rates at the different positions, especially at sites G_1 and G_2. For example, annual N_2O emission rates measured at G_1 in 2002 were 42.8 kg N ha^{-1} y^{-1} at G_{1t} and 11.2 kg N ha^{-1} y^{-1} at G_{1b}.

Annual NO emission rates calculated from the measured NO fluxes ranged from -0.1 to 1.8 kg N ha^{-1} y^{-1}(Table 3). The average of these values was 0.28 kg N ha^{-1} y^{-1} ($n = 46$) and the background NO emission rate was 0.02 kg N ha^{-1} y^{-1} ($n = 15$; excluding site F_2, which received some inputs in the form of excreta). Here, the background NO emission rate was again defined as the average measured at forest sites with no N input during the research period. As was found for N_2O, significantly higher annual NO emission rates ($P < 0.05$) were observed in the cornfields, but unlike for N_2O, the emission rates were significantly higher for the grassland and pasture sites than for the forest (Table 3).

Regression models for estimating N_2O emission rates

When all monitoring data for annual N_2O emission rates were included in the analysis (E_{all}), N_2O emissions were positively correlated ($r^2 = 0.29$, $P < 0.01$) with the N input at each site, although the scatter plot showed a large degree of variation (Fig. 11a). The regression equation obtained from this analysis is as follows:

$$E_{\mathrm{all}} = 0.3045 + 0.0789\ N_{\mathrm{in}} \qquad (3)$$

where E_{all} is the result of this analysis for all N_2O emission rates (kg N ha^{-1} y^{-1}) measured at the livestock farm and N_{in} is the N input (kg N ha^{-1} y^{-1}) at each site. We define this regression model as the “all-inclusive” regression model. The regression slope (0.0789) of the model (i.e., the emission factor for this livestock farm) was higher than the IPCC default value (0.01), and it became apparent that the extremely high N_2O emission rates had increased the emission factor. Therefore, we divided the N_2O emission rates into two groups: normal values (= E_{norm}), which represent values lower than the average (= 6.6, $n = 37$), and higher values (= E_{high}), which represent values higher than the average ($n = 9$). Compared with the all-inclusive regression model, we found a stronger positive correlation between E_{norm} and N input ($r^2 = 0.61$, $P < 0.01$; Fig. 11b). The regression model we obtained is as follows:

$$E_{\mathrm{norm}} = 0.3045 + 0.0195\, N_{\mathrm{in}} \qquad (4)$$

We defined this regression model as the “normal” regression model. We found no significant correlation between E_{high} and N input ($r^2 = 0.07$, $P = 0.48$), between E_{high} and N output ($r^2 = 0.00$, $P = 0.95$), or between E_{high} and N surplus ($r^2 = 0.15$, $P = 0.30$). However, we found a strongly positive correlation ($r^2 = 0.96$, $P = 0.02$) between E_{high} measured in the ungrazed fields, which comprised the ungrazed cornfields (E_{highug}, n = 4), and N surplus (Fig. 11c). The regression equation that we obtained is as follows:

$$E_{\mathrm{highug}} = -54.07 + 1.51\, N_{\mathrm{surp}} \qquad (5)$$

where N_{surp} is the N surplus (kg N ha^{-1} y^{-1}) at each site.

We found no significant correlations between the data for E_{high} measured in the grazed fields (E_{highg}, $n = 5$) and N input, N output, or N surplus. The average E_{highg} value is shown in Figure 11c by a dotted line, and the equation is as follows:

$$E_{\mathrm{highg}} = 24.2 \text{ kg N ha}^{-1} \text{ y}^{-1} \qquad (6)$$

Equations (5) and (6) were defined as the “high” regression models.

Probability of the appearance of a high N_2O flux

The normal and high regression models require a probability (p_{f}) that a high N_2O flux will occur. This probability is defined by the following equation, which integrates estimates calculated by these two models into an annual emission estimate:

$$p_{\mathrm{f}} = [n_{\mathrm{high}} / n] \times 100 \qquad (7)$$

where n is the number of flux measurements in each chamber for each land-use type in each year, and n_{high} ($< n$) is the number of these flux measurements that were higher than the average of all measured fluxes in all land-use types over the 5-year period.

This p_{f} value would be adequate for integrating values calculated by the normal and high regression models, because the N_2O emission rates and the maximum N_2O fluxes showed a high and significant positive correlation (Fig. 10). This indicates that the overall N_2O emission rate was strongly influenced by the periods with high N_2O flux. Therefore, the probability of a high N_2O emission occurring can be assumed to be similar to the probability of a high N_2O flux occurring.

The threshold value for high N_2O fluxes that we used to calculate p_f was the average of all measured fluxes, which amounted to 211 μg N m^{-2} h^{-1} ($n = 1605$). On this basis, p_f ranged from 11% to 41% in the cornfields, 6% to 26% in the grasslands, 0% to 10% in the pastures, and 0% to 2% in the forest (Table 4). The p_f value was consistently higher in the cornfields than in the all other land-use types (with the exception of grasslands in 2001.

Positive, but marginally significant or non-significant, correlations ($P = 0.1$ to 0.2) were found between the probability of a high N_2O flux occurring and precipitation during certain periods for each land-use type (Fig. 12). The correlations were calculated for precipitation from May to June ($r^2 = 0.48$, $P = 0.19$) in the cornfields, from June to July ($r^2 = 0.59$, $P = 0.13$) in the grasslands, from May to August ($r^2 = 0.56$, $P = 0.15$) in the pastures, and in August ($r^2 = 0.61$, $P = 0.12$) in the forests.

DISCUSSION

Factors controlling N_2O fluxes

N_2O fluxes showed large temporal variability in this study. High N_2O fluxes were observed mainly after fertilization and precipitation from May to October (Fig. 5). Inorganic fertilizers were applied mainly from May to August, and the largest amount of daily precipitation occurred during this period (Fig. 5). Dobbie *et al.* (1999) reported a positive correlation between N_2O emission rates and rainfall 1 week before to 3 weeks after fertilizer application. A study conducted in central Hokkaido (Kusa *et al.* 2002; Toma *et al.* 2007) reported high N_2O fluxes during the onion harvest season due to a combination of root cutting and precipitation. Koga *et al.* (2004) found high N_2O fluxes from crop fields in the Tokachi region of eastern Hokkaido; they reported that N_2O fluxes were related to the rate of N application and to the amount and timing of rainfall. Kusa et al. (2006) also mentioned that water supply was affected by rainfall, and that the increased water supply must have increased the N_2O emission rates and affected the variability in N_2O fluxes. Thus, N fertilizer application and greater rainfall appeared to enhance N_2O fluxes, and it will be important to consider the relationships among N_2O fluxes, fertilization, and rainfall in future studies.

The high N_2O fluxes at our study site could have resulted from high denitrification activity. The N_2O/NO ratios that we measured ranged from 0.1 to 5511 (Fig. 7) but mostly ranged between 1 and 100, and N_2O flux increased with an increase in this ratio (Fig. 8). Toma and Hatano (2007) and Toma *et al.* (2007) reported similar results. Lipschultz *et al.* (1981) reported that the N_2O/NO ratio ranged from 0.2 to 1.0 during the nitrification process and was approximately 100 during denitrification. Linn and Doran (1984) summarized the relationship between WFPS and microbial activities and reported that nitrification reached a peak when WFPS was between 40% and 60%, and that denitrification reached a peak when WFPS was greater than 60%. The annual average WFPS values for cornfields, grasslands, pastures, and forests in the present study were 61%±9%, 78%±8%, 80y±15%, and 59y±11%, respectively. These high WFPS values suggest that the high N_2O flux resulted from high levels of denitrification. Furthermore, WFPS values at maximum N_2O flux (Fig. 9) were 58% for the cornfields, 56% for the forests, 74% for the grasslands, 72% for the pastures, and 67% for all land-use types combined in the regression curves created by the boundary-line approach. Thus, high N_2O emission rates in the grasslands and pastures are likely to have been caused by high denitrification levels.

Annual N_2O emission rates

The higher N_2O fluxes from the cornfields may have resulted from the slower and incomplete N uptake by plants and the higher denitrification rates, similar to what Bouwman (1996) reported. We could not observe a clear pattern of variation in N uptake for any of the land-use types in this study, but incomplete N uptake was observed in the cornfields (Table 2). The N surplus in cornfields (a mean of 54±28 kg N ha^{-1} y^{-1}) was higher than those of all other land-use types (29±39, 19±34, and -0.04±0.07 kg N ha^{-1} y^{-1} for grasslands, pastures, and forests, respectively). A large variation in the N_2O fluxes at the study site was reported in our previous study (Katayanagi and Hatano 2005). In that study, we measured N_2O fluxes along a line transect at the livestock farm after fertilization and before harvesting, and we reported a large spatial variability in N_2O fluxes (46±105 and 51±140 μg N m^{-2} h^{-1}, respectively). Our present results also showed large spatial variability. N_2O fluxes measured at some positions within a site differed throughout the year, as was the case for G_{1t} and G_{1b} in 2002 (Fig. 5). Corre *et al.* (1996) reported that the measurement of N_2O grouped based on a landform classification would improve the quantification of N_2O emission rates. They indicated that the soil water content was an important factor, and that soil water content must be controlled by topography, which influences more fundamental hydrologic and pedologic processes. The higher N_2O emission rates at G_{1t} than at G_{1b} in 2002 could also have been influenced by topography. However, we could not clearly demonstrate this effect of topography on N_2O emission rates here, but the observed variation suggests that it is sufficiently important to justify further research aimed at reducing the uncertainty in N_2O emission rates that results from topographic variations.

The range of annual N_2O emission rates at the livestock farm was very wide (-1.0 to 80.8 kg N ha^{-1} y^{-1}). Akiyama *et al.* (2006) compiled and analyzed data on N_2O emission rates from agricultural fields (crops, tea, and paddy) and found N_2O emission rates from upland Japanese fields ranging from 0.07 to 23.3 kg N ha^{-1}. Mori *et al.* (2005) measured annual N_2O emission rates in grasslands growing on an Andosol in Tochigi Prefecture, Japan; they ranged from 0.39 to 1.59 kg N ha^{-1}. Annual N_2O emission rates from forests, as measured by Oura *et al.* (2001, 2004), ranged from 0.03 to 0.65 kg N ha^{-1} y^{-1}. In the literature review by Akiyama *et al.* (2006), the highest N_2O emission rates were reported by Kusa *et al.* (2006), who measured values ranging from 7.3 to 23.3 kg N ha^{-1} y^{-1}. Kusa *et al.* (2006) suggested that a difference in soil permeability to water strongly influenced the N_2O emission rate, and that a high N_2O emission rate could occur in Japanese agricultural Andosols when they are poorly drained. Most of our research sites were also located on Andosols with a high WFPS year-round, and where soils were occasionally saturated after heavy precipitation. This characteristic of our site supports the suggestion by Kusa *et al.* (2006).

Regression models for estimating N_2O emission rates

The emission factors obtained from the simple all-inclusive regression method that included even the extremely high N_2O emissions was 0.0789 (Fig. 11a; equation 3). This value was about eight times the default emission factor value (0.01) recommended by IPCC (1997). However, the emission factor values reported by Bouwman (1996), which included all types of crops and vegetation, ranged from 0.000 to 0.078, and those reported by IPCC (2001) ranged from 0.0025 to 0.06. Moreover, the range of emission factors estimated for Japanese crop fields by Kusa *et al.* (2002), Nishimura *et al.* (2005), Akiyama *et al.* (2006), and Toma *et al.* (2007) were 0.011 to 0.064, 0.027 to 0.080, 0.0007 to 0.0330, and -0.052 to 0.091, respectively. Thus, our result is within the upper limits of previously reported emission factor values. When the emission factor was calculated by using only E_{norm} (i.e., discounting the outliers), we obtained an emission factor of 0.0195 (Fig. 11b and equation 4), which was within the range of the reported emission factor values.

Here, we explained N_2O emission rates not only by N inputs at a site, but also by the site's N surplus. Evaluation of N_2O emission rates from the N surplus values has also been reported by Kaiser and Ruser (2000), who reported a stronger correlation between the N_2O emission rate and the N surplus than between the N_2O emission rate and N input. In our results, the high N_2O emission rates from the cornfields were positively correlated with the corresponding N surplus (Fig. 11c). This result indicates that N uptake by corn affected the N_2O emission rates and that the relationship between N_2O emission and N surplus must be taken into consideration when estimating N_2O emission rates.

Estimation methods for N_2O emissions that use emission factors or regression models developed using the values of N inputs and N surplus are useful; however, high N_2O emission rates ("outliers") are generally excluded when emission factors are calculated or when regression models are developed. For example, Bouwman (1996) excluded high N_2O emission rates when calculating emission factors. Similarly, Kaiser and Ruser (2000) obtained regression models by using cluster analysis to exclude these outliers. However, N_2O emission rates vary by orders of magnitude within a sampling site, both spatially and temporally, so the exclusion of outliers can lead to underestimation of N_2O emission. Therefore, high N_2O emission rates, which were strongly associated with high levels of total annual N_2O emission rates, must be included in N_2O estimations.

The approach we proposed here enabled us to include outliers in the N_2O estimation by dividing the measured values into normal and higher-than-average groups and accounting for the probability of occurrence of high values. Inclusion of high N_2O emission rates accounts for the annual variation in N_2O emission rates and thereby reduces the uncertainty of N_2O estimation. Furthermore, the new approach is potentially able to predict future N_2O emission rates by using meteorological data, because the probability of occurrence of high N_2O emission rates showed a weak positive correlation with precipitation. If improved predictive ability can be achieved, a large number of measurements of N_2O emission rates may no longer be required to estimate N_2O emission rates. Therefore, the model and probability approach described here may represent an interesting new N_2O estimation method for watersheds that will contribute to reducing the uncertainty in N_2O estimation. In a future paper (Katayanagi *et al.*, manuscript in preparation), we will compare N_2O estimations calculated by four methods, including the models and probability approach described here, and we will elucidate the advantages of improving the method described here for this watershed.

CONCLUSIONS

N_2O fluxes showed large temporal variability. High N_2O fluxes were produced mainly by denitrification enhanced by N fertilizer applications and high WFPS. Annual N_2O emission rates from the livestock farm were higher than values previously reported in Japan. Annual N_2O emission rates from the cornfield were significantly higher ($P <$

0.05) than those from the other land-use types, which did not differ significantly each ohter. We developed a new model that accounted for the reality of high N_2O emission rates, which are normally excluded from N_2O estimations, by using the N inputs, the N surplus, the probability of occurrence of these high rates, and precipitation. The proposed model is a useful new tool because it can capture the annual variation in N_2O emission rates and thereby reduce some of the uncertainty of N_2O estimation.

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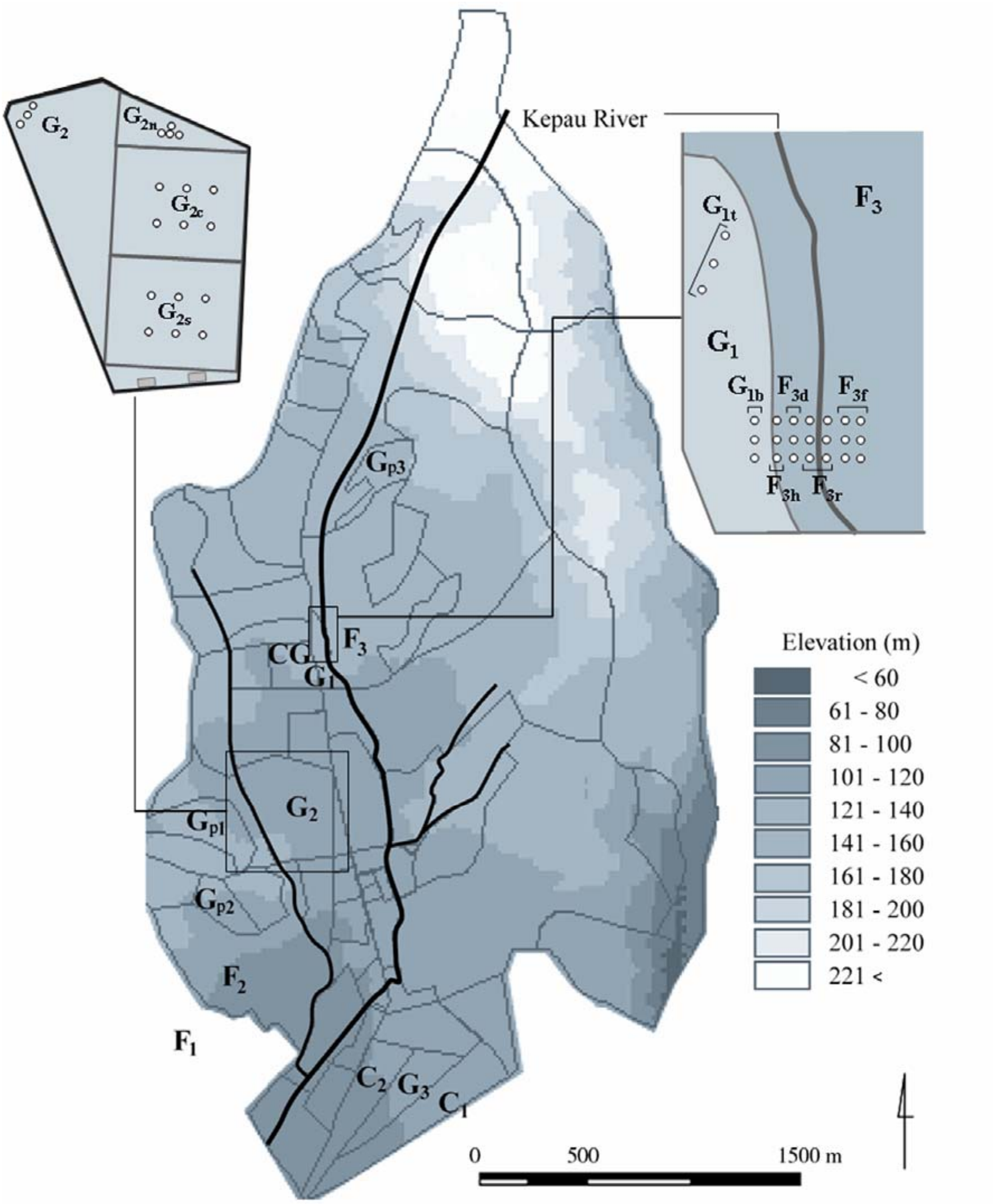

Figure 1. Map of Shizunai Experimental Livestock Farm. The altitude of the farm ranges from 40 to 360 m asl, and the southern and southeastern parts of the farm are lower than the other parts. The monitoring sites included two cornfields (C_1, C_2), four grassland sites (CG [converted from a cornfield in 2003], G_1 [including G_{1t} and G_{1b}], G_2 [including G_{2s}, G_{2c} and G_{2n}], G_3), three pastures (G_{p1}, G_{p2}, G_{p3}), and three forested sites (F_1 [adjacent to the livestock farm], F_2, and F_3 [including F_{3h}, F_{3r}, F_{3d}, and F_{3f}]).

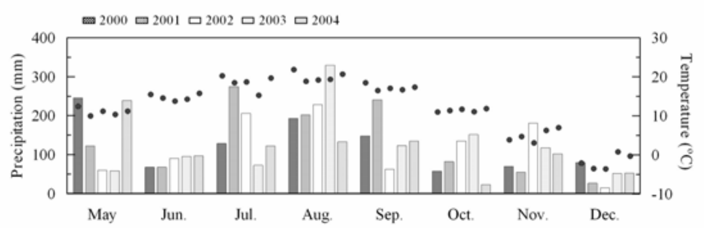

Figure 2. Monthly cumulative precipitation (bars) and mean temperature (circles) from May to December (2000 to 2004). The precipitation data was measured at the Sasayama weather station and the temperatures were recorded at the Shizunai Weather Station.

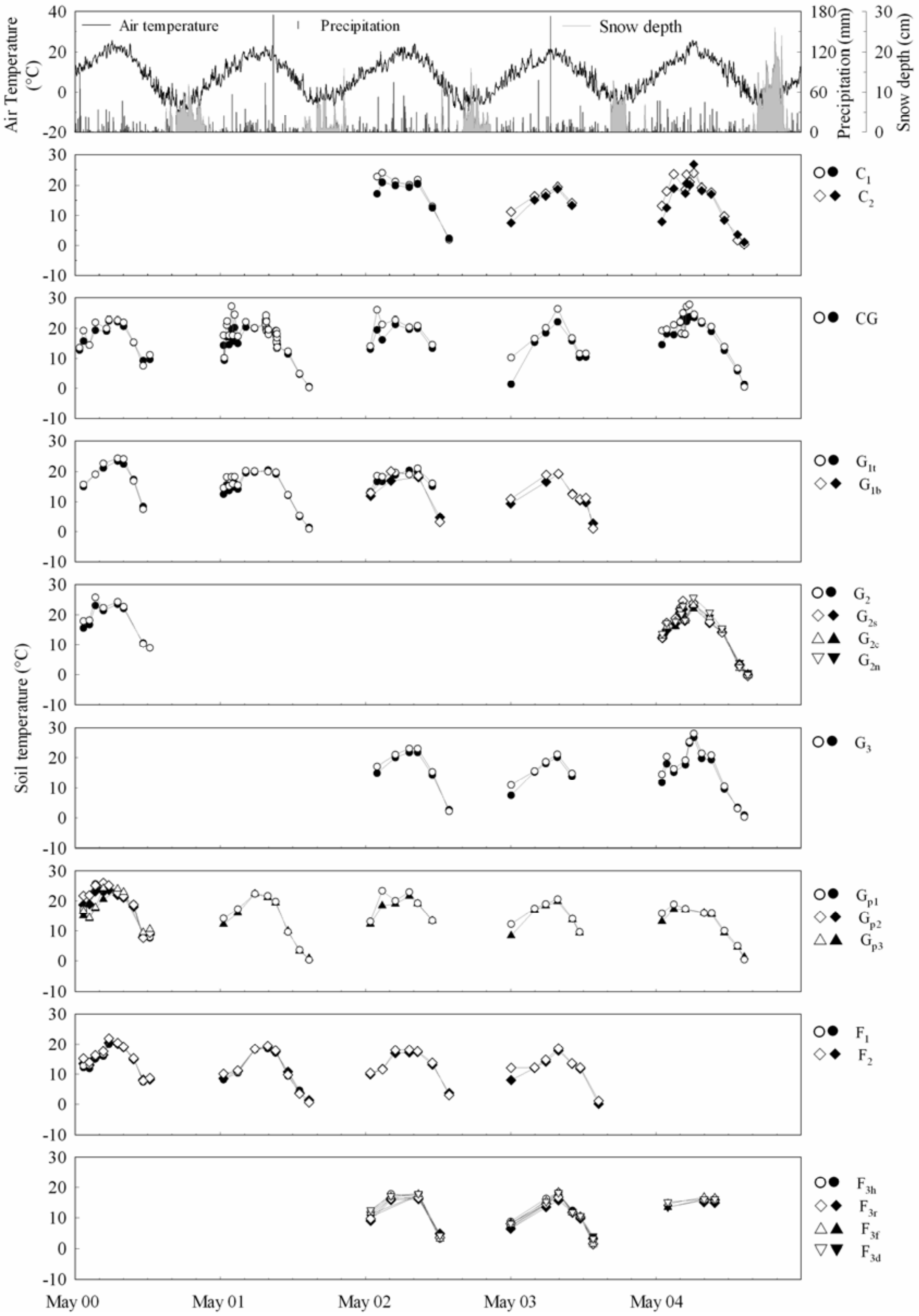

Figure 3. Temporal variability in air temperature, precipitation, snow depth, and soil temperature at depths of 5 cm (o) and 10 cm (•) depth in the cornfields (C_1, C_2), grassland sites (CG, G_{1t}, G_{1b}, G_2, G_{2s}, G_{2c}, G_{2n}, G_3), pastures (G_{p1}, G_{p2}, G_{p3}), and forested sites (F_1, F_2, F_{3h}, F_{3r}, F_{3f}, F_{3d}) at the livestock farm from 2000 to 2004.

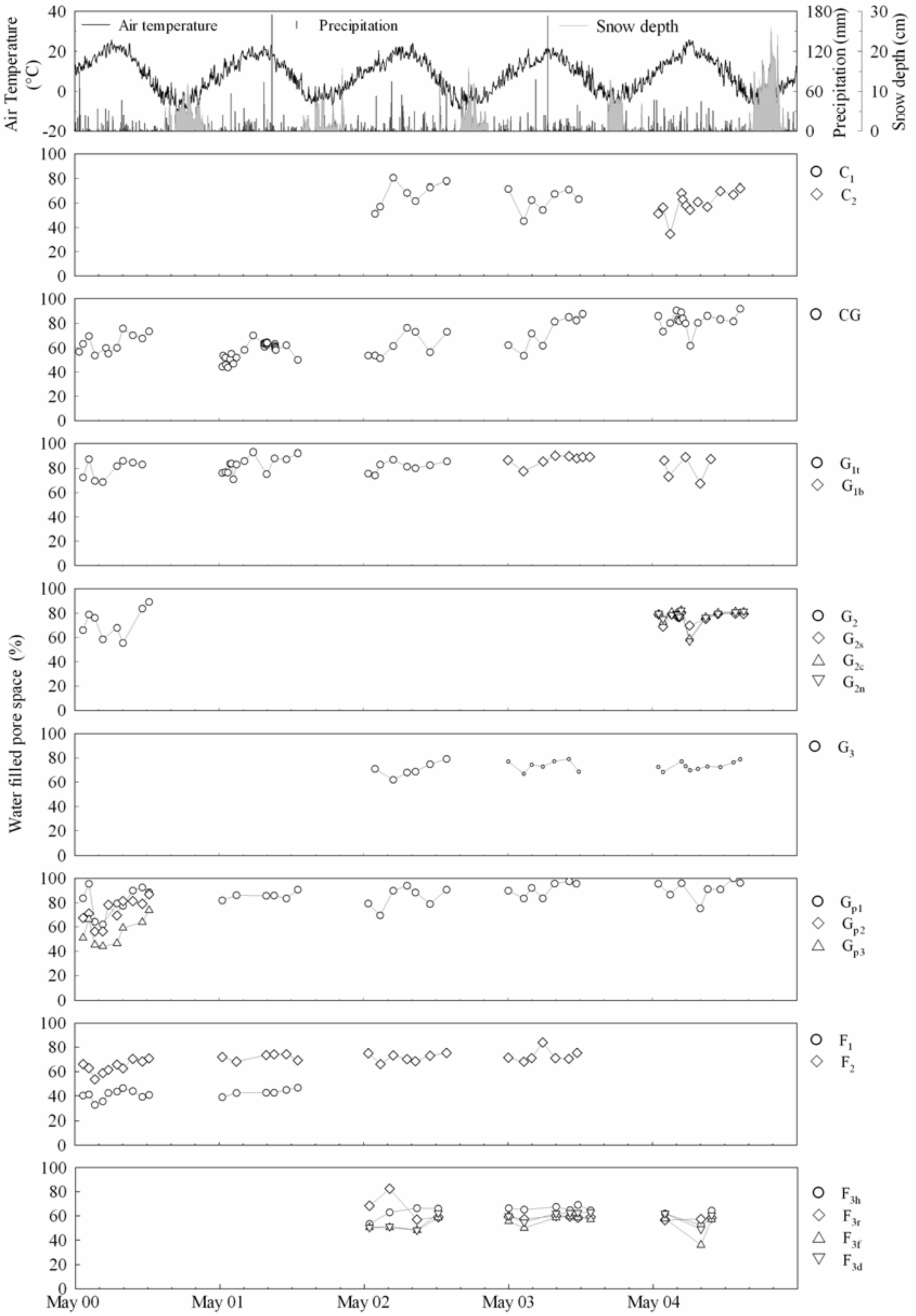

Figure 4. Temporal variability in air temperature, precipitation, snow depth, and water-filled pore space to a depth of 10 cm in the cornfields (C_1, C_2), grassland sites (CG, G_{1t}, G_{1b}, G_2, G_{2s}, G_{2c}, G_{2n}, G_3), pastures (G_{p1}, G_{p2}, G_{p3}), and forested sites (F_1, F_2, F_{3h}, F_{3r}, F_{3f}, F_{3d}) at the livestock farm from 2000 to 2004.

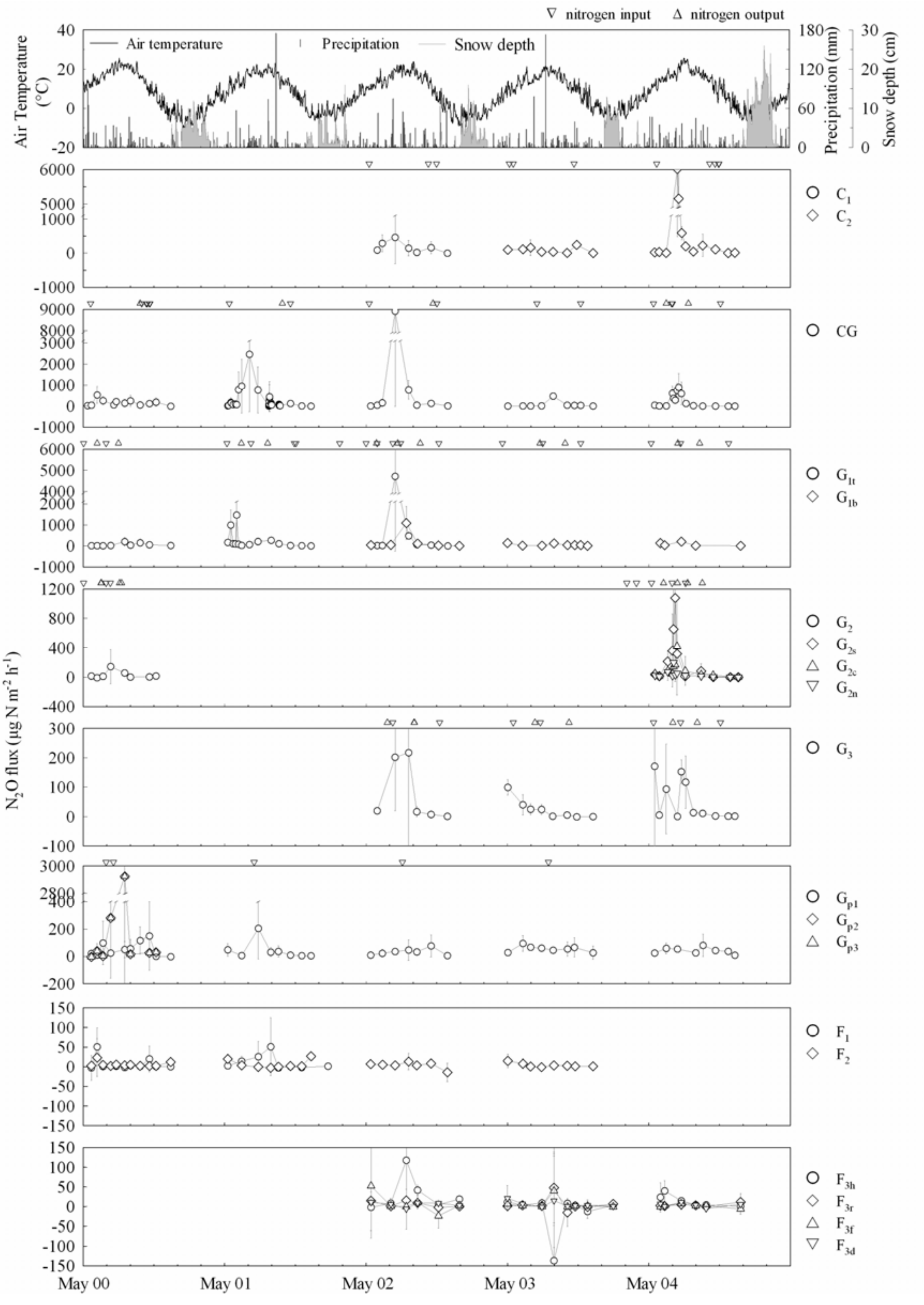

Figure 5. Temporal variability in air temperature, precipitation, snow depth, and N_2O fluxes (mean ± SD) from cornfields (C_1, C_2), grassland sites (CG, G_{1t}, G_{1b}, G_2, G_{2s}, G_{2c}, G_{2n}, G_3), pastures (G_{p1}, G_{p2}, G_{p3}), and forested sites (F_1, F_2, F_{3h}, F_{3r}, F_{3f}, F_{3d}) at the livestock farm from 2000 to 2004. The timing of nitrogen inputs (∇) and outputs (Δ) is shown along the upper edge of each graph. Nitrogen inputs included fertilizer, manure, slurry, and excreta, and nitrogen outputs included yield and feed.

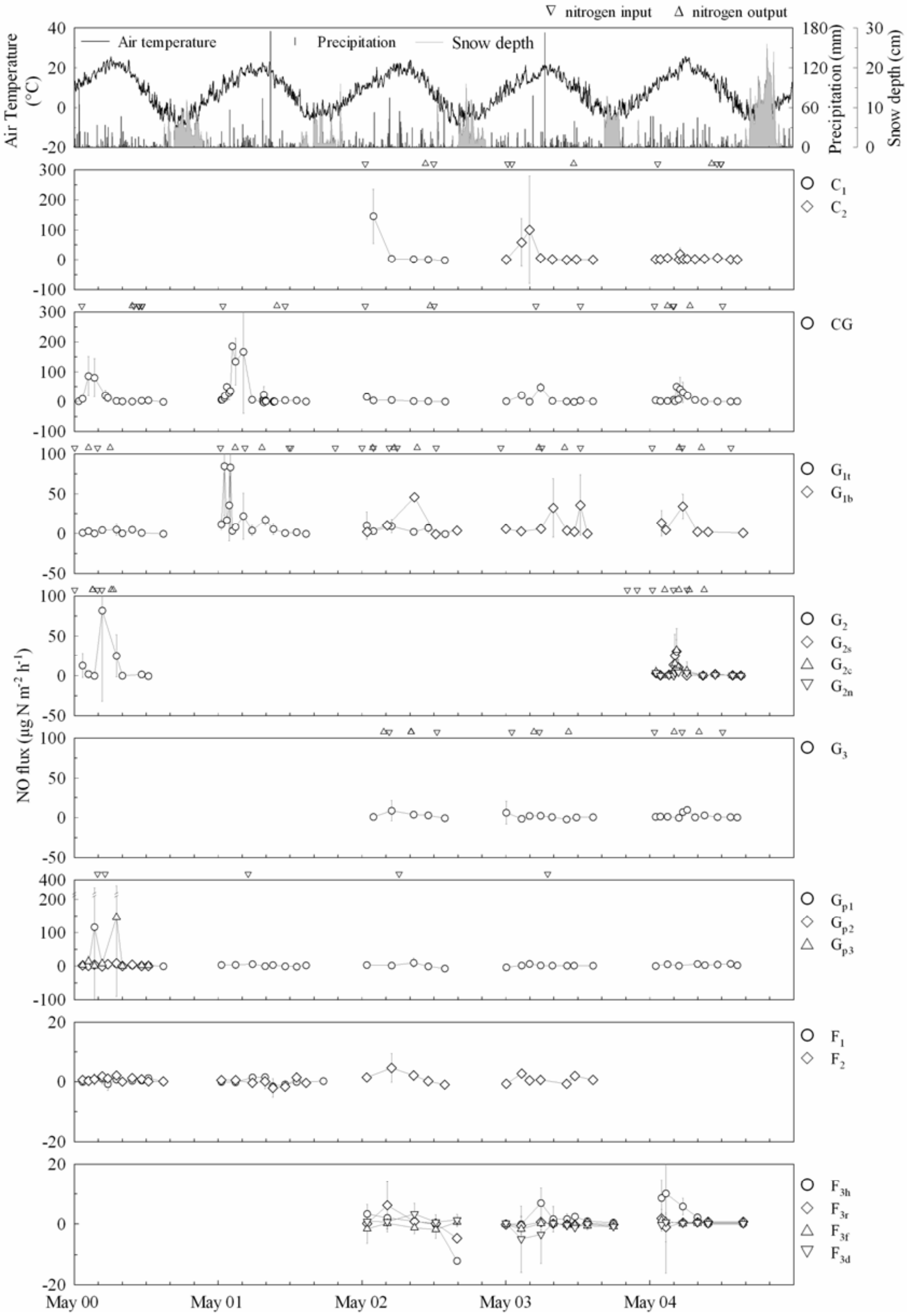

Figure 6. Temporal variability in air temperature, precipitation, snow depth, and NO fluxes (mean ± SD) from cornfields (C_1, C_2), grassland sites (CG, G_{1t}, G_{1b}, G_2, G_{2s}, G_{2c}, G_{2n}, G_3), pastures (G_{p1}, G_{p2}, G_{p3}), and forested sites (F_1, F_2, F_{3h}, F_{3r}, F_{3f}, F_{3d}) at the livestock farm from 2000 to 2004. The timing of nitrogen inputs (∇) and outputs (Δ) is shown along the upper edge of each graph. Nitrogen inputs included fertilizer, manure, slurry, and excreta, and nitrogen outputs included yield and feed.

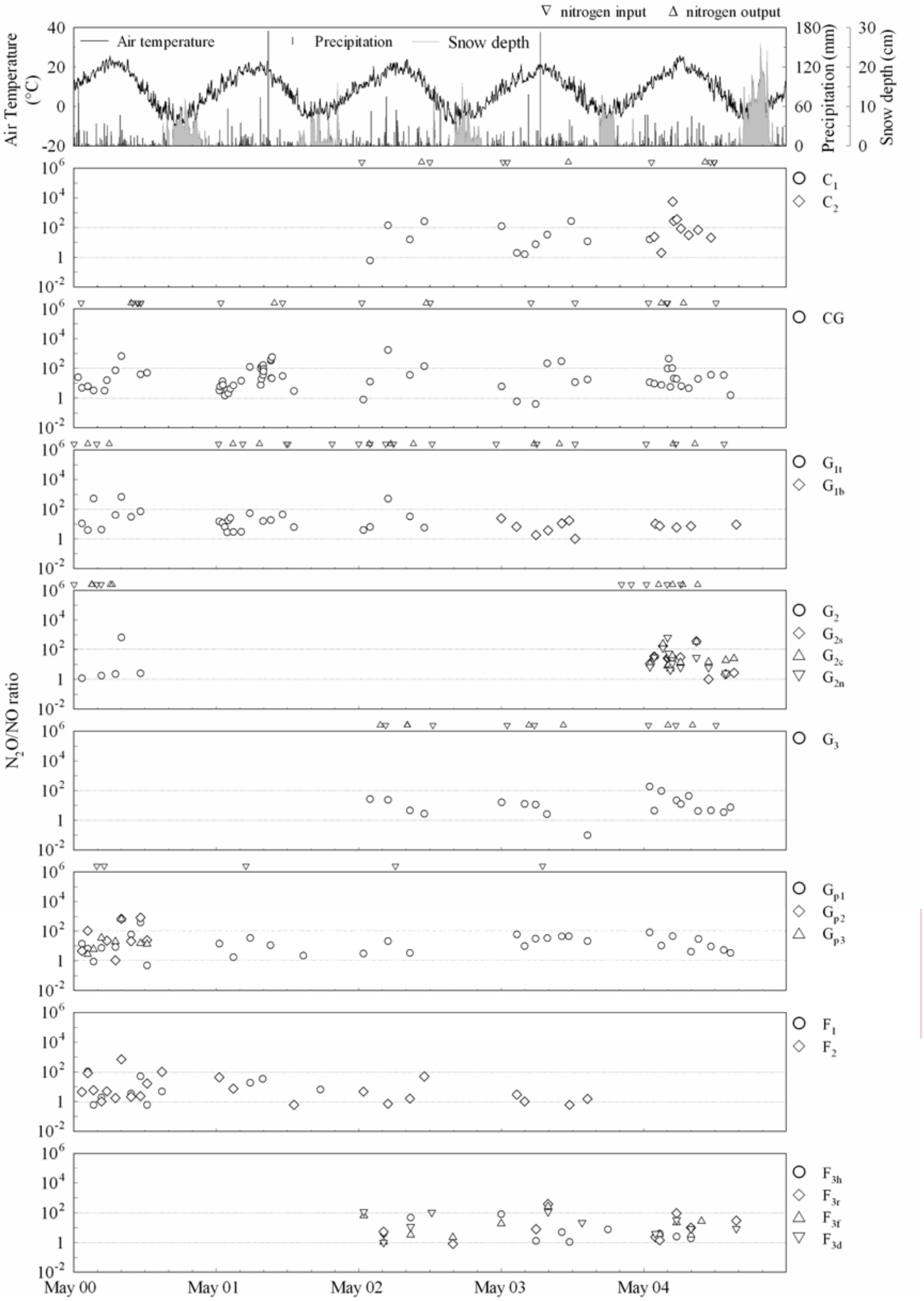

Figure 7. Temporal variability in air temperature, precipitation, snow depth, and the N_2O/NO ratio from cornfields (C_1, C_2), grassland sites (CG, G_{1t}, G_{1b}, G_2, G_{2s}, G_{2c}, G_{2n}, G_3), pastures (G_{p1}, G_{p2}, G_{p3}), and forested sites (F_1, F_2, F_{3h}, F_{3r}, F_{3f}, F_{3d}) at the livestock farm from 2000 to 2004. The timing of nitrogen inputs (∇) and outputs (Δ) is shown along the upper edge of each graph. Nitrogen inputs included fertilizer, manure, slurry, and excreta, and nitrogen outputs included yield and feed.

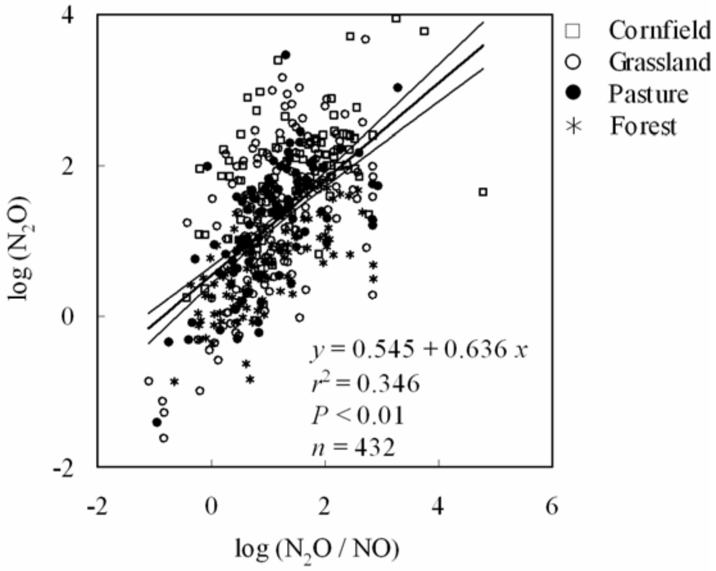

Figure 8. Relationship between log-normal N_2O fluxes (μg N m^{-2} h^{-1}) and the log-normal N_2O/NO ratio of measured values from 2000 to 2004. Bold line represents the regression line, and thin lines represent the two-sided 95% confidence interval.

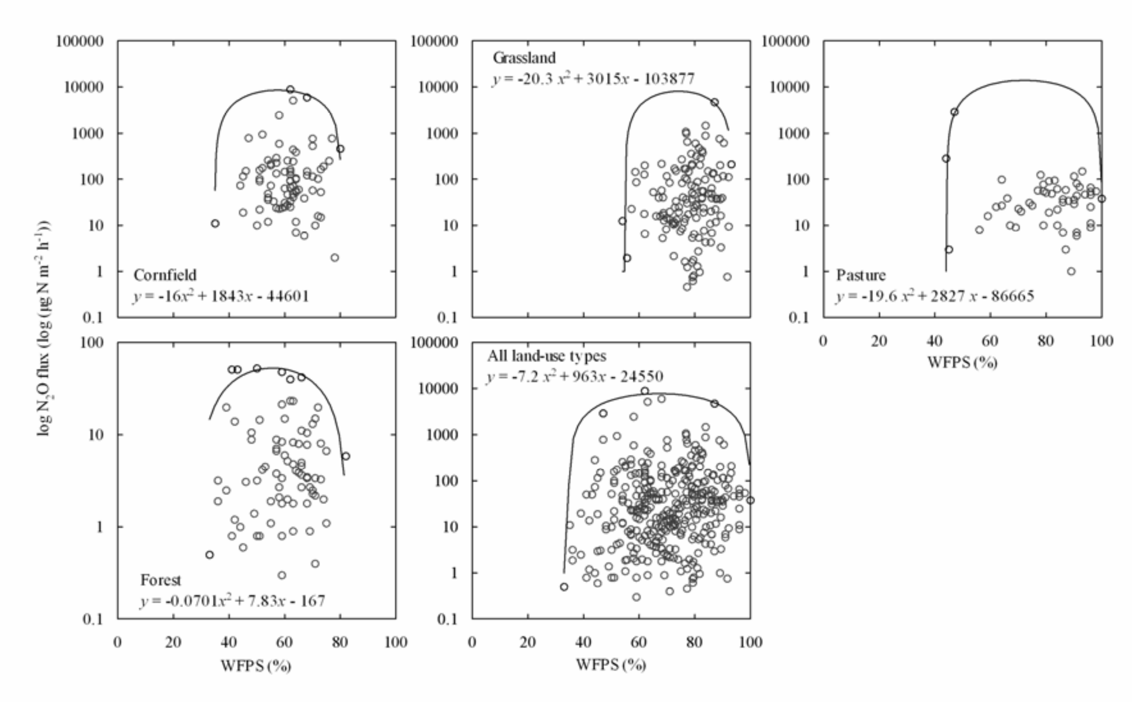

Figure 9. Scatterplot used in the boundary-line analysis for the N_2O emission rate as a function of WFPS (%) for each land-use type (cornfields, grasslands, pastures, and forests) and for all land-use types combined.

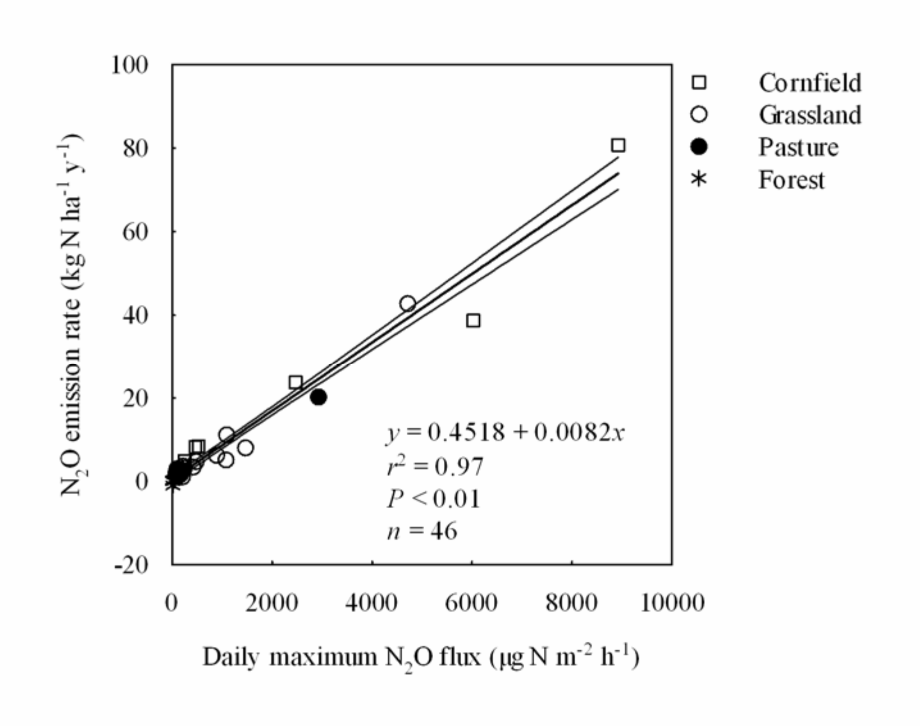

Figure 10. Relationship between maximum N_2O fluxes and annual N_2O emission for all measured annual values from 2000 to 2004. Bold line represents the regression line, and thin lines represent the two-sided 95% confidence interval.

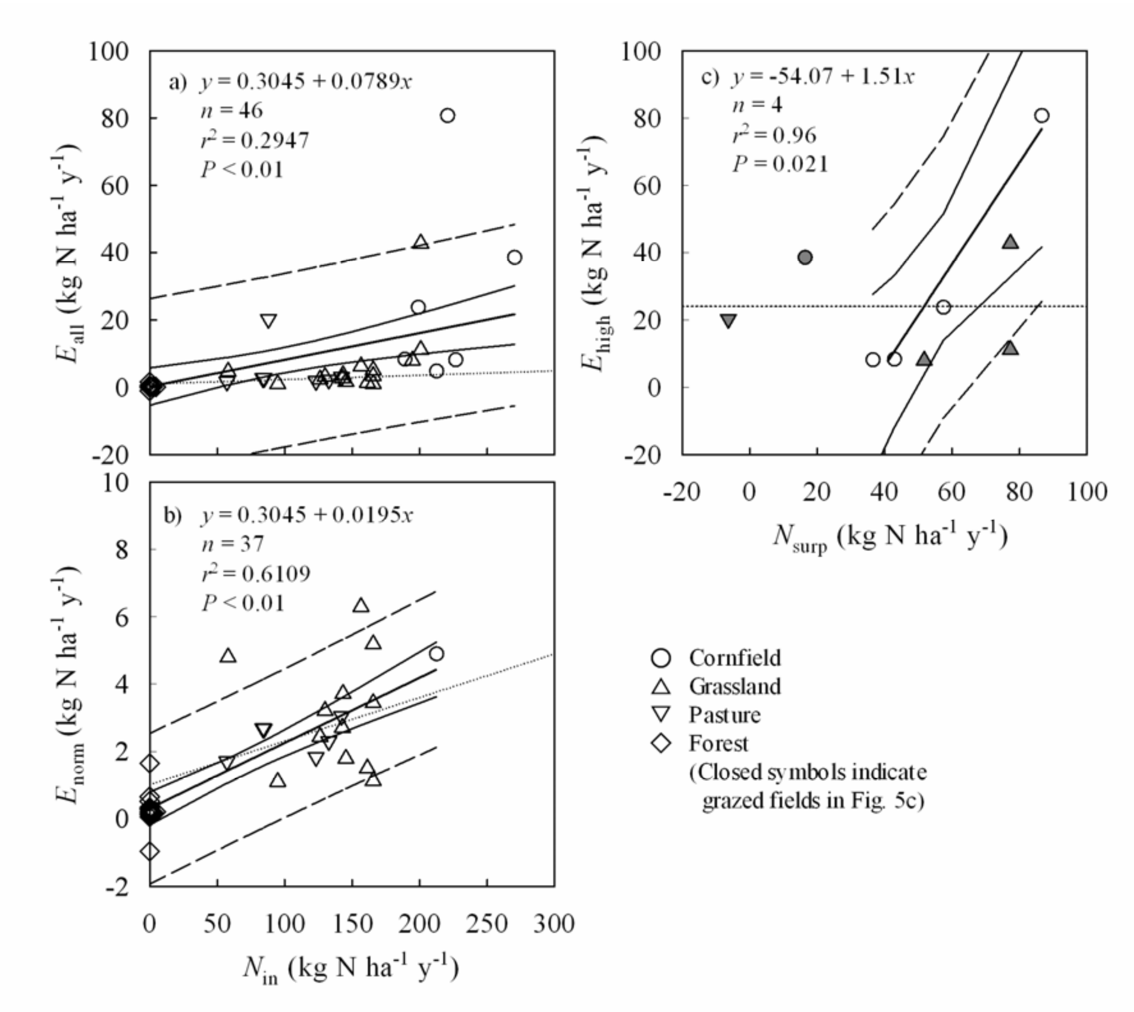

Figure 11. Relationships (a) between E_{all} (n = 46) and nitrogen input, in which all emission values were included; (b) between E_{norm} (n = 37) and nitrogen input, in which only values less than the mean were included; and (c) between E_{high} (n = 9) and nitrogen surplus, in which only values greater than the mean were included, at each site from 2000 to 2004. Solid bold lines represent the regression lines, solid thin lines represent the two-sided 95% confidence interval, and dashed lines represent the 95% prediction interval of the regression line. Dotted lines in (a) and (b) represent values based on the IPCC-recommended model; in (c), the dotted line represents the average value of $E_{\mathrm{highg.}}$

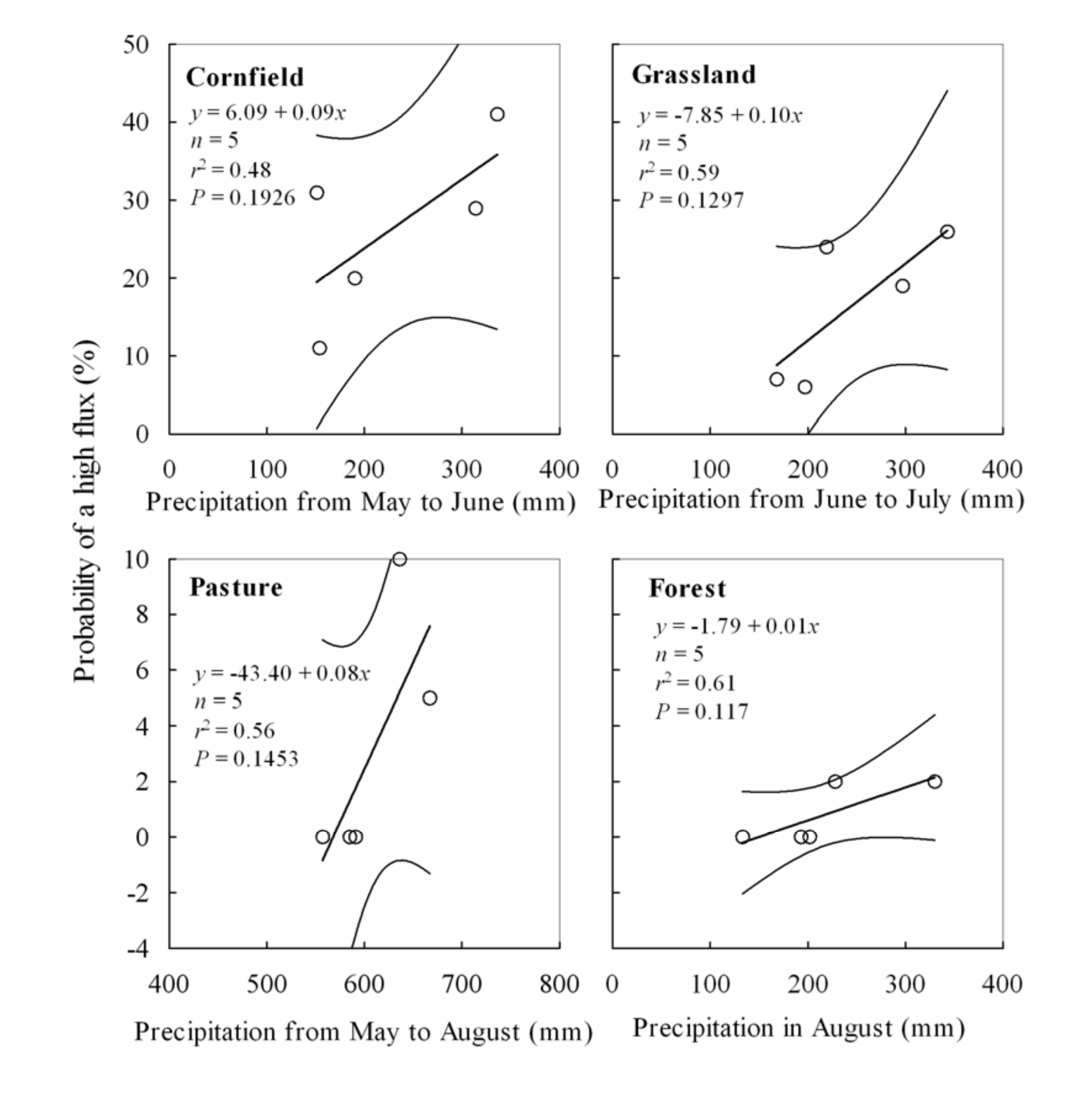

Figure 12. Relationship between the probability of occurrence of a high flux (p_f) for cornfields, grasslands, pastures, and forest as a function of precipitation at the livestock farm from 2000 to 2004. (In this context, a “high” flux represents a flux value greater than the long-term mean for all land-use types.) Solid bold lines represent the regression lines and solid thin lines represent the two-sided 95% confidence interval of the regression lines.

Table 1. Characteristics of the soils in the investigated fields at the Shizunai Experimental Livestock Farm to a depth of 10 cm.

Site	Area (ha)	Research period	Soil classification (FAO)	pH (H_2O)	Total C (g kg^{-1})	Total N (g kg^{-1})	C/N ratio	Bulk density (Mg m^{-3})	Porosity (m m^{-3})
Cornfield									
C_1	5.8	2002-2003	Histosol	5.2	100	6.7	14.9	0.60	0.72
C_2	4.0	2004	Histosol	5.3	60.8	4.5	13.5	0.60	0.72
CG	5.4	2000-2002	Vitric Andosol	5.3	41.8	3.5	12.1	0.78	0.66
Grassland									
CG	5.4	2003-2004	Vitric Andosol	5.3	41.8	3.5	12.1	0.78	0.66
G_{1t}	1.8	2000-2002	Vitric Andosol	6.1	61.7	5.6	11.2	0.71	0.72
G_{1b}	1.8	2002-2004	Vitric Andosol	5.1	50.0	4.2	12.1	0.82	0.67
G_2	7.0	2000	Vitric Andosol	5.2	NA[†]	NA[†]	NA[†]	0.77	0.72
G_{2s}[‡]	1.0	2004	Vitric Andosol	5.1	32.2	3.0	10.6	0.81	0.75
G_{2c}[‡]	1.0	2004	Vitric Andosol	5.0	42.4	3.4	12.4	0.68	0.74
G_{2n}[‡]	0.3	2004	Vitric Andosol	5.3	49.7	4.3	11.5	0.63	0.74
G_3	3.8	2002-2004	Histosol	5.6	75.2	5.4	14.0	0.65	0.72
Pasture									
G_{p1}	2.4	2000-2004	Vitric Andosol	5.7	53.3	4.2	12.6	0.86	0.69
G_{p2}	2.0	2000	Vitric Andosol	5.9	28.6	2.6	11.2	1.10	0.61
G_{p3}	3.1	2000	Vitric Andosol	5.6	29.5	2.3	12.7	0.81	0.72
Forest									
F_1	-	2000-2001	Vitric Andosol	5.7	80.8	6.1	13.2	0.43	0.84
F_2	21.1	2000-2003	Vitric Andosol	6.1	93.6	7.0	13.4	0.55	0.75
F_3	11.2	2002-2004	Vitric Andosol	6.1	73.6	5.7	12.8	0.68	0.71

[†] Not analyzed, NA

[‡] G_2 was divided into G_{2s}, G_{2c}, and G_{2n} in 2004.

Table 2. Nitrogen inputs, output, and surplus (input minus output) in each field where N_2O fluxes were measured at the study site from 2000 to 2004.

Site	Area	Year	Land-use	N input (kg N ha^{-1} y^{-1})					N output (kg N ha^{-1} y^{-1})			N surplus
	(ha)		type†	Fertilizer	Manure	Slurry	Excreta	*Total*	Feed	Yield	*Total*	(kg N ha^{-1} y^{-1})
C_1	5.8	2002	C	153	74	0	0	227	0	190	190	37
		2003	C	126	87	0	0	212	0	129	129	84
C_2	4.0	2004	C	130	123	9.0	8.9	270	12	242	254	16
CG	5.4	2000	C	89	86	14	0	189	0	146	146	43
		2001	C	105	94	0	0	199	0	142	142	57
		2002	C	132	89	0	0	221	0	134	134	87
		2003	G	40	18	0	0	58	0	0	0	58
		2004	G	103	38	16	0	157	0	58	58	99
G_1	1.8	2000	G	99	0	0	31	130	33	88	120	9.5
		2001	G	104	43	0	47	195	51	92	143	52
		2002	G	89	19	90	3.0	201	3.1	121	124	77
		2003	G	84	32	0	10	126	11	75	85	41
		2004	G	75	55	0	12	143	17	101	118	25
G_2	7.0	2000	G	105	0	25	32	161	34	103	137	24
		2004	G	102	0	63	0	166	0	142	142	24
G_3	3.8	2002	G	95	36	0	12	143	13	136	148	-5.1
		2003	G	95	0	0	0	95	0	137	137	-42
		2004	G	103	43	0	0	145	0	157	157	-12
G_{p1}	2.4	2000	P	25	0	0	59	85	63	0	63	21
		2001	P	21	0	0	63	84	68	0	68	15
		2002	P	18	0	0	105	123	113	0	113	10
		2003	P	46	0	0	96	142	104	0	104	38
		2004	P	0	0	0	133	133	150	0	150	-17

† Corn field: C; Grassland: G; Pasture; P; Forest: F

Table 2. (Continued) Nitrogen inputs, N output, and surplus (input minus output) in each field where N_2O fluxes were measured at the study site from 2000 to 2004.

Site	Area (ha)	Year	Land-use type[†]	N input (kg N ha^{-1} y^{-1}) Fertilizer	Manure	Slurry	Excreta	*Total*	N output (kg N ha^{-1} y^{-1}) Feed	Yield	*Total*	N surplus (kg N ha^{-1} y^{-1})
G_{p2}	2.0	2000	P	35	0	0	22	57	24	0	24	33
G_{p3}	3.1	2000	P	0	0	0	88	88	94	0	94	-6.4
F_1	-	2000	F	0	0	0	0	0	0	0	0	0
		2001	F	0	0	0	0	0	0	0	0	0
F_2	21.1	2000	F	0	0	0	0	0	0	0	0	0
		2001	F	0	0	0	2.4	2.4	2.6	0	2.6	-0.2
		2002	F	0	0	0	4.5	4.5	4.6	0	4.6	-0.1
		2003	F	0	0	0	1.0	1.0	1.1	0	1.1	-0.1
F_3	11.2	2002	F	0	0	0	0	0	0	0	0	0
		2003	F	0	0	0	0	0	0	0	0	0
		2004	F	0	0	0	0	0	0	0	0	0

[†] Corn field: C; Grassland: G; Pasture; P; Forest: F

Table 3. Mean, range, standard deviation (SD), and number of measurements (*n*) of N_2O and NO emissions from 2000 to 2004.

Year	Land-use type	N_2O emission (kg N ha^{-1} y^{-1})					NO emission (kg N ha^{-1} y^{-1})				
		n	Mean[†]	Min.	Max.	SD[‡]	*n*	Mean[†]	Min.	Max.	SD[‡]
2000	Cornfield	1	8.5	8.5	8.5	-	1	0.9	0.9	0.9	-
	Grassland	2	2.3	1.5	3.2	1.2	2	0.4	0.1	0.7	0.5
	Pasture	3	8.3	1.7	20.3	10.5	3	0.6	0.1	1.0	0.5
	Forest	2	0.3	0.3	0.3	0.0	2	0.0	0.0	0.0	0.0
2001	Cornfield	1	23.8	23.8	23.8	-	1	1.8	1.8	1.8	-
	Grassland	1	8.0	8.0	8.0	-	1	0.6	0.6	0.6	-
	Pasture	1	2.7	2.7	2.7	-	1	0.1	0.1	0.1	-
	Forest	2	0.4	0.2	0.7	0.3	2	0.0	0.0	0.0	0.0
2002	Cornfield	2	44.5	8.3	80.8	51.3	2	0.5	0.1	0.9	0.5
	Grassland	3	19.3	3.7	42.8	20.8	3	0.4	0.2	0.8	0.4
	Pasture	1	1.8	1.8	1.8	-	1	0.1	0.1	0.1	-
	Forest	5	0.5	0.1	1.7	0.6	5	0.0	0.0	0.1	0.1
2003	Cornfield	1	4.9	4.9	4.9	-	1	1.0	1.0	1.0	-
	Grassland	3	2.8	1.1	4.8	1.9	3	0.4	0.0	0.6	0.3
	Pasture	1	3.0	3.0	3.0	-	1	0.1	0.1	0.1	-
	Forest	5	0.0	-1.0	0.3	0.6	5	0.0	-0.1	0.1	0.1
2004	Cornfield	1	38.7	38.7	38.7	-	1	0.3	0.3	0.3	-
	Grassland	6	3.4	1.1	6.3	2.0	6	0.2	0.1	0.4	0.1
	Pasture	1	2.3	2.3	2.3	-	1	0.2	0.2	0.2	-
	Forest	4	0.2	0.1	0.5	0.2	4	0.0	0.0	0.1	0.1
Mean	Cornfield	6	27.5^{a}	4.9	80.8	29.1	6	0.8^{a}	0.1	1.8	0.6
	Grassland	15	6.6^{b}	1.1	42.8	10.4	15	0.3^{b}	0.0	0.8	0.3
	Pasture	7	4.9^{b}	1.7	20.3	6.8	7	$0.3^{b, c}$	0.1	1.0	0.4
	Forest	18	0.3^{b}	-1.0	1.7	0.5	18	0.03^{c}	-0.1	0.1	0.1

[†] Overall mean represent the 5-year mean of the annual gas emission for each land-use type; in contrast, the overall minimum represents the lowest value during the study period and the overall maximum represents the highest value. Values followed by different letters differed significantly different according to the Tukey test ($P < 0.05$).

[‡] “-” was used in a cell when the standard deviation could not be calculated.

Table 4. Probability of a high N_2O flux occurring (p_f) for each land-use type from 2000 to 2004.

Land-use type	p_f[†] (%)				
	2000	2001	2002	2003	2004
Cornfield	29	20	31	11	41
Grassland	6.4	26	19	7.3	24
Pasture	10	4.8	0.0	0.0	0.0
Forest	0.0	0.0	1.8	2.0	0.0
All land-use types	3.5	3.6	3.4	2.2	2.8

[†] p_f is the mean probability weighted by the area of the given land-use type.