

**RUNNING HEAD:**

New method for estimating N<sub>2</sub>O emission

**TITLE:**

A new method for estimation of nitrous oxide emission rates from an agricultural watershed

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**ABSTRACT**

We developed a new and improved method, named as ‘high-emission-incorporation (HEI) method’, for estimating soil nitrous oxide (N<sub>2</sub>O) emission rates at a watershed level, based on nitrogen (N) input (consisting of fertilizer, manure, slurry and excreta N) to and N surplus (calculated by subtracting the amount of crop yield and consumed N from the N input) of different sites in a livestock farm located in a watershed. The main characteristic of this method is the inclusion of extremely high N<sub>2</sub>O emission rates, called “outlier”, which is normally excluded from estimation. High N<sub>2</sub>O emission rates were estimated using the regression model obtained from the measured N<sub>2</sub>O values and the amounts of N surplus as well as normal N<sub>2</sub>O emission rates were estimated using the regression model obtained from the measured values and the amount of N input. The probability of occurrence of a high flux was used to incorporate calculated high and normal N<sub>2</sub>O emissions into one.

The annual N<sub>2</sub>O emission rates from the livestock farm in the watershed (467 ha), estimated using the HEI method, was 1156±147 kg N y<sup>-1</sup> in a 5-year period. Whereas, the annual N<sub>2</sub>O emission rates calculated using the site-specific emission factor (EF=0.0789) and the emission factor of the Intergovernmental Panel on Climate Change (EF=0.01) was 1838±585 kg N y<sup>-1</sup> and 673 (522 to 1103) kg N y<sup>-1</sup>, respectively. The estimated value using the measure-and-multiply method, in which each land-use area multiplied the representative emission rate for each land-use type, was 964 (509 to 1610) kg N y<sup>-1</sup>. N<sub>2</sub>O emission rates estimated by our newly developed method were consistent with the value calculated by the measure-and-multiply method and offered improvement over it, as it could also predict the future N<sub>2</sub>O emission rates from the

watershed.

**Key words:** emission factor, high-emission-incorporation method, measure-and-multiply method, outlier, uncertainty.

## INTRODUCTION

Nitrous oxide (N<sub>2</sub>O) is a major greenhouse gas and soil is one of its main sources. The global N<sub>2</sub>O emission rate from soils has been estimated to be 10.2 Tg N y<sup>-1</sup>. This is 58% of the total N<sub>2</sub>O emission rate of 17.7 Tg N y<sup>-1</sup> (Intergovernmental Panel on Climate Change 2001). Agricultural fields are especially considered to be one of the major sources of N<sub>2</sub>O. However, because of the large uncertainty in temporal and spatial variability in N<sub>2</sub>O fluxes, it is difficult to estimate an accurate N<sub>2</sub>O emission rate.

The use of emission factors (EFs) is one of the N<sub>2</sub>O estimation method recommended by the United Nations Framework Convention on Climate Change (UNFCCC). The Intergovernmental Panel on Climate Change (IPCC) proposed the EF and the N<sub>2</sub>O estimation method that uses the EFs is named the IPCC Tier 1 method (Intergovernmental Panel on Climate Change 1997 and 2006). When using the IPCC Tier 1 method, N<sub>2</sub>O emission rates are estimated using the sum of the background emission rate and the emission rates resulting from the N inputs to a field. This is done by multiplying the N input by the EF. Actually, the default EF value recommended by the Intergovernmental Panel on Climate Change (1997) is 0.0125, which was calculated by Bouwman (1996). This value was based on the results of only 20 experiments that did not include leguminous crop data and was assumed to include large uncertainty (Bouwman 1996). The default EF was changed from 0.0125 (the uncertainty range =0.0025-0.06) in IPCC (1997) to 0.01 (the uncertainty range =0.003-0.03) in IPCC (2006) based on some reports (e.g. Bouwman *et al.* 2002a, Bouwman *et al.* 2002b, Novoa and Tejeda 2006, Stehfest and Bouwman 2006). In addition to those reports, some other studies also have reported different EF values (e.g., 0.0048 by Kaiser & Ruser 2000; 0.0118 and 0.002 by Helgason *et al.* 2005; 0.011 to 0.064 by Kusa *et al.* 2002; 0.013 to 0.055 by Toma *et al.* 2007) and it indicates that there is large variability in EFs and that EFs still have to be measured to estimate more exact N<sub>2</sub>O emission rates.

Another major N<sub>2</sub>O estimation method is the measure-and-multiply method on field monitoring data (Schimel & Potter 1995; Reiners *et al.* 1998; Corre *et al.* 1999). In this method, the N<sub>2</sub>O emission rate is calculated by multiplying the representative values obtained from field measurements by the surface area of the vegetation, soil, ecosystem, or biome. The measure-and-multiply method is simple and is commonly used, because the method requires a few measured values for each surface type and it is easy to extrapolate those values to a wide area with an assumption that the N<sub>2</sub>O fluxes of the investigated site shows the typical N<sub>2</sub>O emission of that surface type through the study region (Corre *et al.* 1999). The advantage of this method is that the uncertainty caused by the difference among surface types can be decreased by collecting representative values for each surface type, though many samples would be required if emission factors were calculated for each surface type using this method. Furthermore,

this method can represent inter-annual variation if the representative values of N<sub>2</sub>O emission rates were measured at each surface type every year though estimated values calculated by the IPCC tier 1 method cannot represent it. Inter-annual variation in N<sub>2</sub>O emission is very high and measurements would be required if one tried to represent and evaluate the variation (e.g. Toma *et al.* 2007).

The measure-and-multiply method is a more advanced method than the method using EFs, but the problem of excluded extremely high N<sub>2</sub>O emission rates or outliers from analysis should be addressed in both methods. Kaiser and Ruser (2000) defined outliers using the cluster analysis by dividing N<sub>2</sub>O emission rates into three groups: manure-fertilized plots with high N-related N<sub>2</sub>O emissions, legume plots with very low N input-output balances, and forage production plots with high N-fertilizer inputs. The outliers were excluded from the regression analysis between N<sub>2</sub>O emission rates and N input or N input-output balances. Helgason *et al.* (2005) excluded an extremely high N<sub>2</sub>O emission value (44 kg N ha<sup>-1</sup>), which was derived from a barley field that received 60 kg of N fertilizer ha<sup>-1</sup> where one site consistently showed emission rates 5–30 times higher than those in other sites in apparently uniform field plot experiments.

In our previous study (Katayanagi *et al.* 2008), results of a 5-year period measurement of N<sub>2</sub>O emission rates of cornfield, grassland, pasture and forest in a livestock farm located in an agricultural watershed were evaluated. We began by first obtaining an all-inclusive regression model, by incorporating all monitoring data on annual N<sub>2</sub>O emission rates at a livestock farm in a watershed and the N input to the soil for each measured site is as follows:

$$E_{\text{all}} = 0.3045 + 0.0789N_{\text{in}} \quad (1)$$

where  $E_{\text{all}}$  is all N<sub>2</sub>O emission rates (kg N ha<sup>-1</sup> y<sup>-1</sup>) measured at the livestock farm and  $N_{\text{in}}$  is the N input (kg N ha<sup>-1</sup> y<sup>-1</sup>) at each site.

However, since our measured annual N<sub>2</sub>O emission data showed a large variation, and N<sub>2</sub>O emission rates from the livestock farm estimated by using the equation (1) included large uncertainty (Katayanagi *et al.* 2008), we divided all N<sub>2</sub>O emission values ( $E_{\text{all}}$ ) into two groups: high values ( $E_{\text{high}}$ , representing values higher than the overall average, 6.6 kg N ha<sup>-1</sup>,  $n=9$ ) and normal values ( $E_{\text{norm}}$ , representing values lower than the overall average,  $n=37$ ). We then developed regression models for  $E_{\text{high}}$  and  $E_{\text{norm}}$ . We defined the equation for  $E_{\text{norm}}$  as the “normal” regression model:

$$E_{\text{norm}} = 0.3045 + 0.0195N_{\text{in}} \quad (2)$$

A significantly positive correlation ( $r^2 = 0.96$ ,  $P < 0.02$ ) was found between  $E_{\text{high}}$  measured in the fields where no grazing had taken place ( $E_{\text{highug}}$ ,  $n=4$ ) and N surplus. However, there was no significant correlation between  $E_{\text{high}}$  measured in the grazed fields ( $E_{\text{highg}}$ ,  $n = 5$ ) and the N input, N output or N surplus. The average  $E_{\text{highg}}$  value was 24.2 kg N ha<sup>-1</sup> y<sup>-1</sup>, and we defined this value as the representative value for high N<sub>2</sub>O emission rates from the grazed field. The obtained regression equations were as follows:

$$E_{\text{highug}} = -54.07 + 1.51N_{\text{surp}} \quad (3)$$

$$E_{\text{highg}} = 24.2 \quad (4)$$

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

We defined equations (3) and (4) as the “high” regression models. Furthermore, we also defined the probability of occurrence of high  $\text{N}_2\text{O}$  emissions ( $p_f$ , %) to integrate estimates calculated by the use of these two models. The  $p_f$  was calculated by the following equation:

$$p_f = [n_{\text{high}} / n] \times 100 \quad (5)$$

where  $n$  is the number of flux measurements in each chamber for each land-use type in each year and  $n_{\text{high}}$  ( $< n$ ) 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.

The calculated  $p_f$  values for cornfield, grassland, pasture and forest showed a weak positive correlation with precipitation from the months of May to June, from June to July, from May to August, and August, respectively (Katayanagi *et al.* 2008). These models and the probability of occurrence must be useful, because they can represent inter-annual variation in  $\text{N}_2\text{O}$  emission rates and can reduce uncertainty in  $\text{N}_2\text{O}$  estimation by including extremely high  $\text{N}_2\text{O}$  emission rates.

In this paper, we propose a new method, the high-emission-incorporation (HEI) method, for estimating soil  $\text{N}_2\text{O}$  emission rates at a watershed level using the models and the probability of occurrence of high emission rates developed in the previous paper (Katayanagi *et al.* 2008). The main characteristic of this method is the inclusion of extremely high  $\text{N}_2\text{O}$  emission rates by using not only the N input, but also the N surplus of a site and meteorological data. To verify the new method, we compared the results calculated by the new method and three other methods: the all-inclusive regression method, the measure-and-multiply method and the IPCC Tier 1 method.

## MATERIALS AND METHODS

### Study area and estimation of the N budget

The study was carried out at the Shizunai Experimental Livestock Farm (467 ha), Field Science Center for Northern Biosphere, Hokkaido University, Japan ( $42^{\circ}25'9''\text{N}$ ,  $142^{\circ}29'1''\text{E}$ ). The detailed field information was described in our previous paper (Katayanagi *et al.* 2008).

Annual N budgets for each site were estimated for a 5-year period from 2000 to 2004 following the method applied by Hayakawa *et al.* (2004). The N input, output and surplus were calculated mainly based on the livestock farm register. While estimating N budgets, the quantity, the variety and timing of application of chemical fertilizer, the compost manure, slurry and feed supply, grazing and harvests of grass were taken into consideration. Ammonia volatilization, denitrification, N fixation and N deposition were not taken into account in the estimation. The detailed methods and calculated results were described in Hayakawa *et al.* (2004) and Katayanagi & Hatano (2005). The

calculated results for the annual N budget of the livestock farm from 2000 to 2004 are summarized in Table 1. During the study period, the total N input ranged from 20,754 to 22,561 kg N y<sup>-1</sup>, the total N output from 13,453 to 19,089 kg N y<sup>-1</sup> and the total N surplus from 1665 to 9108 kg N y<sup>-1</sup>.

### Estimation of annual N<sub>2</sub>O emission rates using the four methods

To verify the total annual N<sub>2</sub>O emission rates from the entire area of the livestock farm located in the watershed using the HEI method, the emission rates were also estimated using three other methods commonly in use such as the all-inclusive method, the measure-and-multiply method (Schimel & Potter 1995; Reiners *et al.* 1998; Corre *et al.* 1999) and the IPCC Tier 1 method (Bouwman 1996; Intergovernmental Panel on Climate Change 1997 and 2006). All these four methods are explained in the following sections and are summarized in Figure 1.

#### *The all-inclusive regression method*

The annual N<sub>2</sub>O emission rate from the watershed based on the all-inclusive method,  $E_{ai}$  (kg N y<sup>-1</sup>), was calculated by using the equation (1) and the following equation:

$$E_{ai} = \sum_{i=1}^n E_{f1i} \cdot A_{fi} \quad (6)$$

where  $n$  is the number of sites;  $E_{f1}$  is the annual N<sub>2</sub>O emission rates (kg N ha<sup>-1</sup> y<sup>-1</sup>) from the sites where N<sub>2</sub>O fluxes were measured and the sites that were not measured. These were also used for the annual N<sub>2</sub>O emission rates from each site which was calculated using the N input into each site and the equation (1);  $A_f$  is the area (ha) of each site. The regression models and 95% confidence interval of equation (1) are given in Figure 2. The confidence interval (95%) of  $E_{ai}$  was calculated by using the standard error of  $E_{f1i}$  and the propagation of error. Uncertainty in  $A_{fi}$  was assumed to be zero.

#### *The high-emission-incorporation method*

The annual N<sub>2</sub>O emission rate from the watershed based on the HEI method,  $E_{hei}$  (kg N y<sup>-1</sup>), was calculated using the following equation:

$$E_{hei} = \sum_{i=1}^n \sum_{j=1}^m [E_{f2i} [1 - (p_{fj} / 100)] + E_{f3i} (p_{fj} / 100)] \cdot A_{fi} \quad (7)$$

where  $n$  is the number of sites in the livestock farm;  $m$  is the number of land-use types at the farm;  $E_{f2}$  and  $E_{f3}$  (kg N ha<sup>-1</sup> y<sup>-1</sup>) are the annual N<sub>2</sub>O emission rates from each site where N<sub>2</sub>O fluxes were measured and from sites where N<sub>2</sub>O fluxes were not measured.  $E_{f2}$  was calculated using the N input into each site and the equation (2) and  $E_{f3}$  was calculated using the N surplus of each site and the equation (3) and the equation

(4);  $p_f$  (%) is the probability of occurrence of high N<sub>2</sub>O emissions calculated by the equation (5); and  $A_f$  is the area (ha) of each site, respectively.  $E_{f2}$  was calculated by using the N input and the equation (2) to estimate normal N<sub>2</sub>O emission rates and  $E_{f3}$  was calculated by the N surplus and equations (3) and (4) to estimate high N<sub>2</sub>O emission rates. A constant value of 8.0 kg N ha<sup>-1</sup> y<sup>-1</sup> was used when the value calculated using the high regression model was lower than the least measured value of  $E_{\text{highug}}$  (= 8.0 kg N ha<sup>-1</sup> y<sup>-1</sup>) for fields that had not been used for grazing. A constant value of 24.2 kg N ha<sup>-1</sup> y<sup>-1</sup> (=  $E_{\text{highg}}$ ), the representative value for a high N<sub>2</sub>O emission rate, was used for the grazed field.  $A_f$  is the area (ha) of each site. The regression models, constants and 95% confidence interval of equation (2), (3) and (4) are shown in Figure 2. A confidence interval (95 %) of  $E_{\text{nei}}$  was calculated following the same method of  $E_{\text{ai}}$ .

#### *The measure-and-multiply method*

The annual N<sub>2</sub>O emission rate from the watershed based on the measure-and-multiply method,  $E_{\text{mm}}$  (kg N y<sup>-1</sup>), was calculated using the following equation:

$$E_{\text{mm}} = \sum_{i=1}^n \sum_{j=1}^m E_{l_j} \cdot A_{f_i} \quad (8)$$

where  $n$  and  $m$  are the number of sites and land-use types at the livestock farm, respectively and  $E_l$  is the average of N<sub>2</sub>O emission rates (kg N ha<sup>-1</sup> y<sup>-1</sup>) from each land-use type in each year. The average values of measured annual N<sub>2</sub>O emission rates were 4.9-80.8 for cornfield, 1.1-42.8 for grassland, 1.7-20.3 for grazing pasture and -1.0-1.7 for forests (Katayanagi *et al.* 2008); these were used to calculate the annual N<sub>2</sub>O emission rate from the watershed.  $A_f$  is the area (ha) of each site where N<sub>2</sub>O fluxes were measured and also of the sites that were not measured. A confidence interval (95 %) of  $E_{\text{mm}}$  could not be calculated, because there were some land-use types where replication was less than two. Therefore, the range of  $E_{\text{mm}}$  for each year was shown in the results.

#### *The IPCC Tier 1 method*

The annual N<sub>2</sub>O emission rate from the watershed based on the IPCC Tier 1 method,  $E_{\text{ipcc}}$  (kg N y<sup>-1</sup>), was calculated by the following equation:

$$E_{\text{ipcc}} = \sum_{i=1}^n E_{f4_i} \cdot A_{f_i} \quad (9)$$

where  $E_{f4}$  is the annual N<sub>2</sub>O emission rate (kg N ha<sup>-1</sup> y<sup>-1</sup>) from the sites where N<sub>2</sub>O fluxes were measured and also of the sites that were not measured. This was calculated using the N input values (kg N ha<sup>-1</sup> y<sup>-1</sup>) for each site and the IPCC-recommended

equation ( $\text{N}_2\text{O}$  emission rate =  $0.01[\text{N input}] + 1$ ; Intergovernmental Panel on Climate Change 2006).  $A_f$  is the area (ha) of each site. The range of  $E_{\text{ipcc}}$  was also calculated for each year by using 0.03% EF for the lower limit and 3% EF for the higher limit. This is as proposed by the Intergovernmental Panel on Climate Change (2006) for uncertainty in EF. In the IPCC Tier 1 method, the  $\text{N}_2\text{O}$  emission rate from crop residue and fixed N by leguminous crops must be estimated. However, we were unable to estimate them for this paper, because there was no crop residue input. Except for corn stubble in the cornfields, there were only a few leguminous types of grass in the grassland and pastures.

### Statistical analyses

Significant differences between  $\text{N}_2\text{O}$  emission rates calculated using the different methods were evaluated by Tukey tests ( $P < 0.05$ ).

## RESULTS

The annual  $\text{N}_2\text{O}$  emission rates from each site, calculated by using the normal and high regression models ( $E_{f2}$  and  $E_{f3}$ ) for  $E_{\text{hei}}$ , ranged from 0.3 to 40.4 kg N ha<sup>-1</sup> y<sup>-1</sup> and from 8.0 to 562 kg N ha<sup>-1</sup> y<sup>-1</sup>, respectively (Table 2). The  $p_f$  for cornfield, grassland, pasture and forest was 11-41%, 6.4-26%, 0-10% and 0-2.0%, respectively (Table 2). The  $p_f$  and its inter-annual variation in cornfield and grassland were very large. The  $p_f$  was 0% for pasture in 2002-2004 and for forest in 2000, 2001 and 2004.

The mean of the annual  $\text{N}_2\text{O}$  emission rates from the watershed for a 5-year period, calculated by using the all-inclusive regression method ( $E_{\text{ai}}$ ), HEI method ( $E_{\text{hei}}$ ), measure-and-multiply method ( $E_{\text{mm}}$ ) and the IPCC Tier 1 method ( $E_{\text{ipcc}}$ ), was 1838, 1156, 964 and 673 kg N y<sup>-1</sup>, respectively (Table 3).  $E_{\text{ai}}$  and  $E_{\text{ipcc}}$  always showed the largest and smallest values, respectively among the values estimated by the four methods.  $E_{\text{hei}}$  and  $E_{\text{mm}}$  indicated similar mean, minimum and maximum values for the 5-year period. However, the pattern of inter-annual variation in  $E_{\text{hei}}$  and  $E_{\text{mm}}$  was not consistent; i.e.  $E_{\text{hei}}$  showed the highest value in 2001, while  $E_{\text{mm}}$  showed the highest value in 2002.

Comparison of the confidence interval between  $E_{\text{ai}}$  and  $E_{\text{hei}}$  showed that the interval for  $E_{\text{ai}}$  was wider than that for  $E_{\text{hei}}$  every year (Table 3). However the inter-annual variation in the confidence interval for  $E_{\text{ai}}$  was smaller than that for  $E_{\text{hei}}$ . Furthermore, the confidence interval for  $E_{\text{mm}}$  and  $E_{\text{ipcc}}$  could not be calculated; because  $E_{\text{mm}}$  did not have enough replications for each land-use type and each year to calculate a confidence interval. A confidence interval for the IPCC Tier 1 method was not given in IPCC (2006). The inter-annual variations in minimum and maximum values of  $E_{\text{ipcc}}$  were very small, whereas those of  $E_{\text{mm}}$  were very large (Table 3).

## DISCUSSION

The annual  $\text{N}_2\text{O}$  emission rates from the watershed calculated by the all-inclusive regression method,  $E_{\text{ai}}$ , were significantly higher than those by the other methods (Table 3), and this result was caused by the higher site specific EF (=0.0789,

see equation 1) based on the N input values rather than the IPCC recommended EF. The reason for the high site specific EF of the all-inclusive regression method was because the N<sub>2</sub>O emission rates were much higher than the normal values, i.e. “outliers” and were included in the calculations of EF. The large variation in N<sub>2</sub>O emission rates in space and time were reported by many researchers (e.g. Velthof & Oenema 1995; Katayanagi & Hatano 2005) and the existence of “outliers” has been reported in previous papers (e.g. Kaiser & Ruser 2000; Helgason *et al.* 2005). Therefore, to reduce uncertainty in N<sub>2</sub>O estimation, it would be better to include outliers by using a different method.

Although the inter-annual variation and the confidence interval in  $E_{ai}$  were smaller than in  $E_{hei}$ ,  $E_{ai}$  was significantly higher than  $E_{hei}$  (Table 3). The reason for the small inter-annual variation in  $E_{ai}$  was because the estimates calculated by using a site specific EF for a 5-year period could only represent the inter-annual variation in N input rates. The IPCC Tier 1 method also had such limitations similar to the all-inclusive regression method.  $E_{ipcc}$  showed the smallest estimates (Table 3) because the EF of the IPCC Tier 1 method was smaller than that of the all-inclusive regression method. In fact, the inter-annual variation in N input to each site was smaller than that of the N<sub>2</sub>O emission rate (coefficient of variation for N input was 3%; Table 1), whereas that for N<sub>2</sub>O emission rates calculated by the measure-and-multiply method was 36% in a 5-year period (Table 3). To solve the problems concerning the method using an EF to estimate N<sub>2</sub>O emission which can not represent the inter-annual variation in N<sub>2</sub>O emission rates and to reduce the uncertainty in N<sub>2</sub>O estimation, a site specific EF of each research area for each year should be calculated. However, it would be much more costly to calculate a site specific EF for each research area for every year and it would require a method which enables estimating N<sub>2</sub>O emission rates without continuous monitoring. In such a context, we have developed a new method named the “high-emission-incorporation (HEI) method”.

The HEI method is considered a more advanced method than the method using an EF and has just as much as the measure-and-multiply method. This is because, as explained in the Introduction section,  $E_{mm}$  values obtained by the measure-and-multiply method are considered to be more reliable and is a likely value than the values obtained by a single emission factor ( $E_{ai}$  and  $E_{ipcc}$ ). In fact,  $E_{mm}$  values were substantially different from  $E_{ai}$  and  $E_{ipcc}$  values (Table 3). Furthermore,  $E_{hei}$  based on our new method was similar to  $E_{mm}$  and showed similar inter-annual variation to  $E_{mm}$  (Table 3).

The large inter-annual variation in  $E_{hei}$  was similar to that of  $E_{mm}$ , however, the pattern of inter-annual variation in  $E_{mm}$  and  $E_{hei}$  was different. The reason for this could be that the application methods of incorporating the outlier values differ between the HEI method and the measure-and-multiply method. In the measure-and-multiply method, the representative values were calculated using a few measured N<sub>2</sub>O emission rates. This means, there is a possibility that outliers may not have been taken into consideration, especially in a year that annual N<sub>2</sub>O emission rates were calculated from a few samples. A low  $p_f$  indicates the probability of their neglect, because the  $p_f$  is the probability of occurrence of high N<sub>2</sub>O emission rates and a low  $p_f$  means that the probability of occurrence of high N<sub>2</sub>O emission rates in a few replications is low.



On the other hand, the HEI method can include outliers by estimating the probability of occurrence of high emission rates, which are calculated from a number of N<sub>2</sub>O flux values. At this point, it can be said that the HEI method would be a more useful technique compared to the measure-and-multiply method. This is in order to lessen the possibility of overlooking the extreme emission values and the uncertainty in N<sub>2</sub>O estimation.

In addition, the HEI method is useful in predicting inter-annual variation in N<sub>2</sub>O emission rates at a larger scale, because the occurrence probability of high N<sub>2</sub>O emissions showed a positive correlation with cumulative precipitation from May to June in the cornfield, from June to July in the grassland, from May to August in the pasture, and in August in the forest (Katayanagi *et al.* 2008). N<sub>2</sub>O emission rates are also reported to increase after fertilization and precipitation (Katayanagi *et al.* 2008). Some reports indicated the relationship between water-filled pore spaces (WFPS), which is influenced by precipitation, and N<sub>2</sub>O emission rates (Kusa *et al.* 2006; Koga *et al.* 2004). When the timing of the N application was different among the land-use types, the WFPS values were also different in the case of the highest N<sub>2</sub>O emission rates (Katayanagi *et al.* 2008). Therefore, it could also be possible that the  $p_f$  for each land-use type showed a positive correlation with the different accumulation period of precipitation.

The HEI method is a newly developed method and must be useful for N<sub>2</sub>O estimation in a watershed scale, but the method leaves room for improvement in some fields such as adequacy of threshold value to divide the overall flux values into higher and normal fluxes or emission rates. This also applies to unclear mechanisms for estimating the probability of occurrence of high emission rates and consideration of other soil types and climatic conditions.

The HEI method is new and improved in terms of estimating much more reliable N<sub>2</sub>O emission rates at a watershed level including outliers (the extremely higher N<sub>2</sub>O emission rates than the normal values). This method accounted for the existence of outliers by using N inputs, N surplus, and the probability of occurrence of these high rates and precipitation together. It is expected that this new method could become more useful to estimate N<sub>2</sub>O emission rates on a watershed scale by incorporation of some of these improvements.

## CONCLUSION

The high-emission-incorporation method, in which normal N<sub>2</sub>O emission rates and higher N<sub>2</sub>O emission rates than the normal values were calculated and incorporated into one by using the probability of occurrence of a high flux, improved the estimation of N<sub>2</sub>O emission rates. The N<sub>2</sub>O emission rates from the watershed calculated using this method were consistent to the estimates based on the measure-and-multiply method. It also could predict the future N<sub>2</sub>O emission rates from the watershed by using the input and surplus N, and precipitation.

## ACKNOWLEDGEMENTS

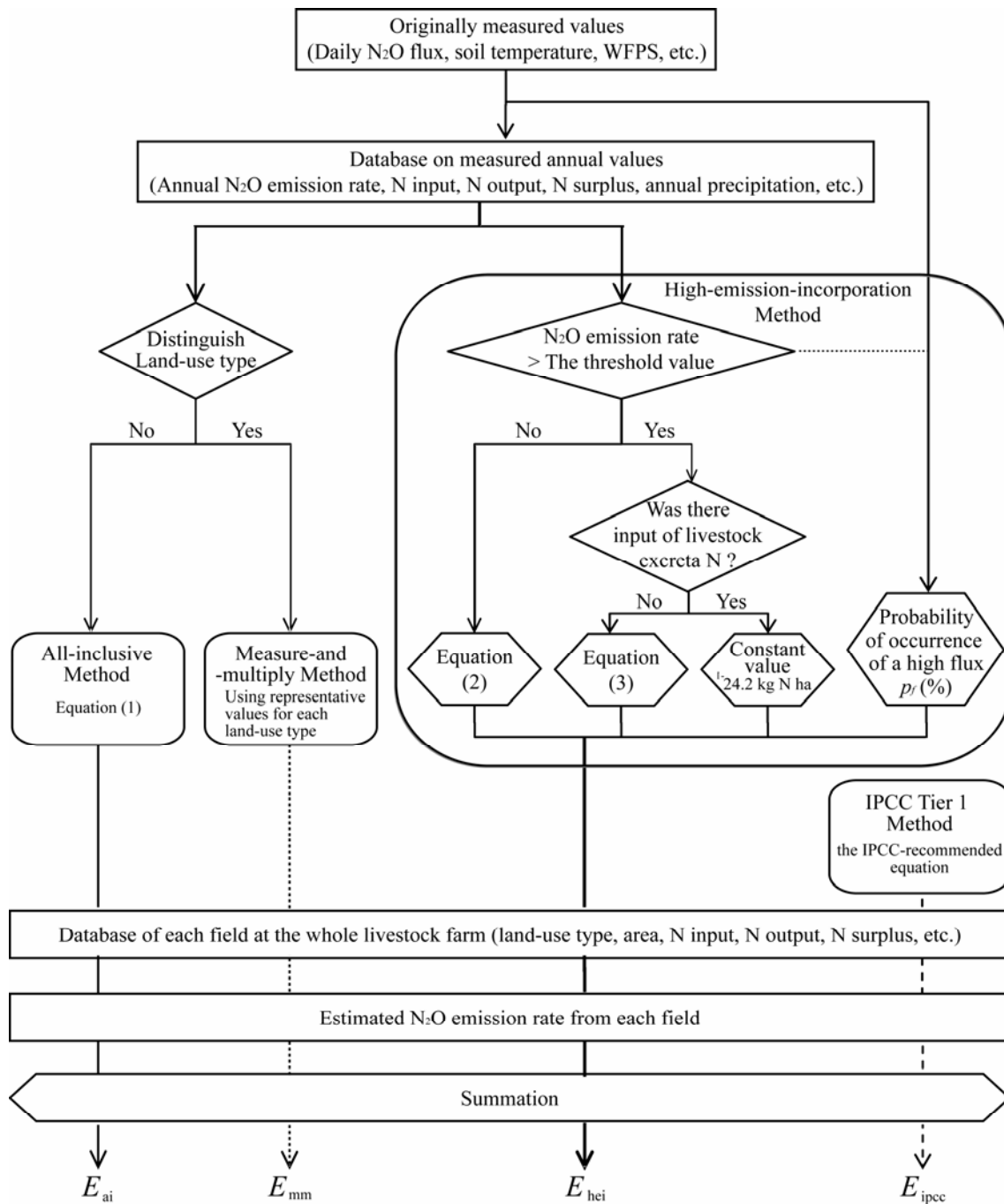
We thank Dr. H. Hata and the staff of Shizunai Experimental Livestock Farm,

Field Science Center, Hokkaido University for their assistance; Dr. O. Nakahara of the Hokkaido University, Dr. K.P. Woli of the University of Illinois and Dr. S. D. Kimura of the Tokyo University of Agriculture and Technology for useful discussions; and Miss M. Takemoto of Oyo Corporation, Miss M. Shimizu of the Soil Science Laboratory, Mr. S. Okada of Asahi Breweries, LTD. and Miss Y. Usui of New Energy and Industrial Technology Development Organization for their assistance in sampling. This paper was presented at the International Workshop on Monsoon Asia Agricultural Greenhouse Gas Emissions (MAGE-WS), March 7-9, 2006, Tsukuba, Japan. This study was partly supported by a Japanese Grant-in-Aid for Science Research from the Ministry of Education, Culture, Sports, Science and Technology (No.11460028).

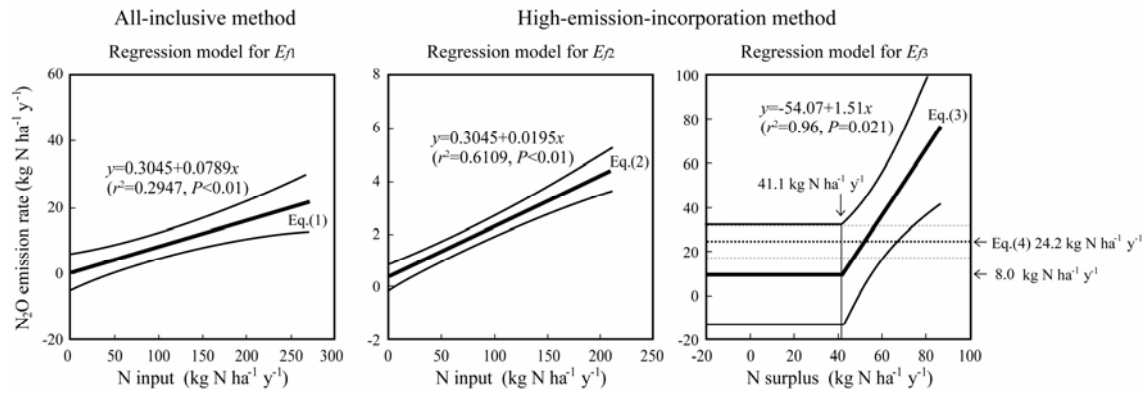
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**Figure 1.** A schematic diagram of different approaches for scaling up the estimation of annual  $\text{N}_2\text{O}$  emission rates from the site to the watershed.



**Figure 2.** A diagram of the regression models for  $E_{f1}$ ,  $E_{f2}$  and  $E_{f3}$ . The solid bold lines represent the regression lines obtained for calculating  $E_{f1}$ ,  $E_{f2}$  and  $E_{f3}$ . The solid thin lines represent the regression lines at a 95% confidence interval and the dotted bold line represents  $E_{f4}$ . The dotted line represents the standard error of  $E_{f4}$ .

**Table 1.** N input to, N output from, and surplus N (kg N y<sup>-1</sup>) at the livestock farm (467 ha) in a watershed in 2000-2004

Year	2000	2001	2002	2003	2004	Mean	SD	CV, %
N input	21226	22561	20754	21646	21228	21483	680	3.2
N output	15464	13453	19089	13544	15854	15481	2293	15
N surplus	5762	9108	1665	8102	5374	6002	2886	48

**Table 2.**  $E_{f2}$  and  $E_{f3}$  of each land-use type in the livestock farm in 2000-2004

Year	Land-use type	$n$	$E_{f2}$ (kg N ha <sup>-1</sup> y <sup>-1</sup> )				$E_{f3}$ (kg N ha <sup>-1</sup> y <sup>-1</sup> )				$P_f^\dagger$ (%)
			Mean	Min	Max	SD	Mean	Min	Max	SD	
2000	Cornfield	2	3.4	2.7	4.0	0.9	9.3	8.0	10.7	1.9	29
	Grassland	10	3.6	2.6	5.7	0.9	42.2	8.0	124	42.9	6.4
	Pasture	36	3.9	0.9	25.8	5.2	32.6	24.2	208	35.8	10
	Forest	12	0.4	0.3	0.5	0.1	16.1	8.0	24.2	8.5	0.0
	<i>Total</i>	60	3.1	0.3	25.8	4.3	30.1	8.0	208	33.7	3.5
2001	Cornfield	2	8.3	4.2	12.4	5.8	297	32.7	561	374	20
	Grassland	10	3.5	2.5	4.1	0.5	39.5	8.0	141	42.9	26
	Pasture	36	3.6	1.2	35.4	5.6	32.1	24.2	309	47.4	4.8
	Forest	12	0.3	0.3	0.4	0.0	16.1	8.0	24.2	8.5	0.0
	<i>Total</i>	60	3.1	0.3	35.4	4.7	39.0	8.0	561	79.9	3.6
2002	Cornfield	2	4.2	3.7	4.6	0.6	42.4	8.0	76.8	48.6	31
	Grassland	10	3.9	2.7	7.0	1.3	17.7	8.0	24.2	8.4	19
	Pasture	37	3.2	1.1	14.6	2.4	42.4	8.0	445	82.6	0.0
	Forest	12	0.3	0.3	0.4	0.0	18.8	8.0	24.2	8.0	1.8
	<i>Total</i>	61	2.8	0.3	14.6	2.3	33.7	8.0	445	65.5	3.4
2003	Cornfield	2	6.1	4.4	7.8	2.4	169	72.3	266	137	11
	Grassland	10	2.8	1.4	4.6	0.9	36.4	8.0	151	43.1	7.3
	Pasture	36	4.1	0.3	40.4	6.4	38.3	8.0	562	89.9	0.0
	Forest	12	0.3	0.3	0.4	0.0	20.2	8.0	24.2	7.3	2.0
	<i>Total</i>	60	3.2	0.3	40.4	5.2	38.7	8.0	562	77.8	2.2
2004	Cornfield	3	6.5	5.6	7.4	0.9	187	24.2	329	153	41
	Grassland	9	2.7	0.3	3.7	1.1	29.2	8.0	114	34.1	24
	Pasture	36	4.0	1.1	30.8	5.5	24.2	22.1	24.2	0.4	0.0
	Forest	12	0.3	0.3	0.4	0.0	18.8	8.0	24.2	8.0	0.0
	<i>Total</i>	60	3.2	0.3	30.8	4.6	32.0	8.0	329	47.6	2.8

<sup>†</sup> $P_f$  in the total is the mean weighted by the land area.

**Table 3.** Estimated annual N<sub>2</sub>O emission rates from the farm calculated by the all-inclusive ( $E_{ai}$ ), high-emission-incorporation( $E_{hei}$ ), measure-and-multiply( $E_{mm}$ ), and IPCC Tier 1 ( $E_{ipcc}$ ) methods for each land-use type in 2000-2004

Year	$E_{ai}$ (kg N y <sup>-1</sup> )		$E_{hei}$ (kg N y <sup>-1</sup> )		$E_{mm}$ (kg N y <sup>-1</sup> )			$E_{ipcc}$ (kg N y <sup>-1</sup> )		
	Mean	95% C.I. †	Mean	95 % C.I. †	Mean	Min	Max	Mean	Min‡	Max‡
2000	1815	581	899	65	1110	416	2373	670	521	1094
2001	1920	596	1675	194	966	896	1050	683	525	1134
2002	1777	582	812	69	1421	421	2969	665	520	1080
2003	1848	582	936	68	478	88	629	674	522	1107
2004	1829	585	1457	146	844	722	1030	672	522	1100
<i>Mean</i> <sup>¶</sup>	1838 <sup>a</sup>	585	1156 <sup>b</sup>	108	964 <sup>b, c</sup>	509	1610	673 <sup>c</sup>	522	1103
<i>SD</i>	53		385		347			6.7		
<i>CV, %</i>	2.9		33		36			0.99		

†C.I., confidence interval; CV, coefficient of variation

‡Minimum and maximum of  $E_{ipcc}$  were calculated by using 0.3 % EF and 3 % EF, which was indicated in IPCC (2006) for uncertainty in EF values.

¶Representative values were the average of a five-year period. Values with different letters are significantly different according to a Tukey-Kramer test ( $P < 0.01$ ).