

# Differences in the Spatial Variability Among CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O Gas Fluxes from an Urban Forest Soil in Japan

Sonoko Dorothea Bellingrath-Kimura, Ayaka Wenhong Kishimoto-Mo,  
Noriko Oura, Seiko Sekikawa, Seichiro Yonemura, Shigeto Sudo,  
Atsushi Hayakawa, Kazunori Minamikawa, Yusuke Takata, Hiroshi Hara

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**Abstract** The spatial variability of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O) fluxes from forest soil with high nitrogen (N) deposition was investigated at a rolling hill region in Japan. Gas fluxes were measured on July 25th and December 5th, 2008 at 100 points within a 100 × 100 m grid. Slope direction and position influenced soil characteristics and site-specific emissions were found. The CO<sub>2</sub> flux showed no topological difference in July, but was significantly lower in December for north-slope with coniferous trees. Spatial dependency of CH<sub>4</sub> fluxes was stronger than that of CO<sub>2</sub> or N<sub>2</sub>O and showed a significantly higher uptake in hill top, and emissions in the valley indicating strong influence of water status. N<sub>2</sub>O fluxes showed no spatial dependency and exhibited high hot spots at different topology in July and December. The high N deposition led to high N<sub>2</sub>O fluxes and emphasized the spatial variability.

**Keywords** Carbon dioxide · Methane · Nitrous oxide · Rolling hill · Spatial variability · Urban forest

## INTRODUCTION

The expansion of urban areas and industry mainly impacts forest ecosystems with increased atmospheric deposition (Hatano et al. 2007). This has a big effect on the nutrient cycle, which is especially obvious when considering greenhouse gas (GHG) emissions from soils such as carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O). Estimation of GHG emission from natural ecosystem is important to calculate the influence of human activity on the global climate change. However, accurate estimation is still difficult due to high temporal and spatial variability in natural ecosystems.

Higher atmospheric deposition enhances plant growth and leads to increased litter fall, providing a greater C source than that of enhanced heterotrophic respiration. CH<sub>4</sub> oxidation is susceptible to increased N availability (Jiang et al. 2010) and its reduction leads to higher CH<sub>4</sub> emission, which is produced at anaerobic micro-spots. N<sub>2</sub>O can be produced by denitrification under anaerobic conditions and by nitrification under aerobic conditions, both of which are enhanced if available N increases (hole-in-the-pipe model, Davidson et al. 2000). The closed-chamber method is often used for GHG measurements. In this method, GHG concentration is periodically sampled in a closed chamber to calculate the efflux from the rate of increase or decrease of the gas concentration. The closed-chamber method is used widely due to its convenience, even though a dynamic chamber has a smaller impact on the internal conditions due to its aeration (Bekku et al. 1995).

In Japanese forests, the annual mean N<sub>2</sub>O emission is  $1.88 \pm \text{SD}1.89 \mu\text{g N m}^{-2} \text{h}^{-1}$ , while the CH<sub>4</sub> uptake is  $2.9\text{--}175 \text{ gC m}^{-2} \text{h}^{-1}$  (Morishita et al. 2007). These estimates are obtained from core monitoring sites all over Japan. However, measurements from those points do not take into account topological differences such as slope direction, micro-elevations, and soil conditions. Moreover, forest topology is highly heterogeneous and complex, and the topography can vary from almost flat surfaces to steep slopes or valleys. Soil moisture varies greatly with slope position, and it is usually lower in the upper part of a slope (Borken and Beese 2005). Furthermore, the spatial distribution of soil temperature and other soil properties changes gradually along a slope (Florinsky et al. 2002).

These factors affect GHG emissions, and high spatial and temporal variability has been recorded (Jia et al. 2003; Konda et al. 2008; Nishina et al. 2009a). Lower slope positions have shown N<sub>2</sub>O emissions that are higher than

those of a shoulder position, which were mainly influenced by the decreasing carbon/nitrogen (CN) ratio along the slope (Nishina et al. 2009a). Another study reported that N<sub>2</sub>O emission was influenced by both soil moisture and organic matter (Florinsky et al. 2004). N<sub>2</sub>O and CH<sub>4</sub> emissions are higher at positions where water gathers or the ground water table is high and anaerobic soil conditions develop (Jungkunst et al. 2008). The CO<sub>2</sub> flux is also related to the ground water table (Jungkunst et al. 2008). The turnover rate increases with tree growth. Detto et al. (2013) found that hydrological networks and associated topographical variation had a large influence on the mean canopy profile height at scales of 20–300 m. The influencing factors are scale dependent (Lark et al. 2004), and analysis of small-scale variability in topography of GHG emission is essential for an understanding of GHG emission from forest soils.

The Tokyo prefecture is known to be one of the highest populated areas in the world. Surprisingly, 40% of the prefecture is covered with forest. The remaining forest niches are often found at complex landscapes with high slope angles (Economic Planning Agency 1969). Soil conditions are highly diverse, which have resulted in high heterogeneity of soil processes. Due to this complexity, the GHG emission from these sub-urban forests is highly uncertain. Thus, a forest in a suburban area of Tokyo with a complex topography was chosen to investigate the influence of topology on the spatial distribution of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O gas fluxes.

## MATERIALS AND METHODS

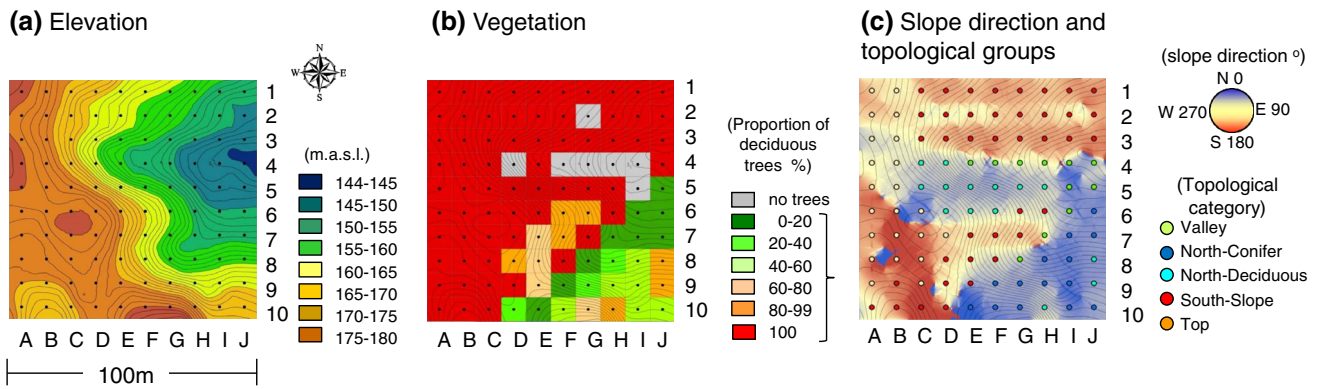
### Study Site

The study area was a site of the Japan Long-Term Ecological Research Network (JaLTER) of associated sites (N35°38′17.62″, E139°22′35.44″) at the Field Museum Tamakyuryo, a field experimental station of the Tokyo University of Agriculture and Technology. The average annual precipitation is 1600 mm and there is an average temperature of 14.4 °C (Japan Meteorological Agency 2010). The station is located at the northeast edge of the Tama Rolling Hills, which spread to the southeast of the Kanto plane containing the highest population density in Japan. Due to its proximity to the city of Tokyo, a large amount of N deposition has been recorded in this area (Kimura et al. 2009). While the average N deposition of Japan ranges from 3.1 to 18.2 kgN ha<sup>-1</sup> y<sup>-1</sup> (Ministry of Environment 2009), in 2007 the open bulk N deposition was 19.7 kgN ha<sup>-1</sup> y<sup>-1</sup> and cedar forest throughfall was 50.6 kgN ha<sup>-1</sup> y<sup>-1</sup> (Kimura et al. 2009).

The JaLTER plot was established in 2006. The plot area was set to a 100 × 100 m grid with sub-plots at 10-m intervals (Fig. 1a). The plot represents the complex topology of the rolling hills and has two valleys. The elevation is in the range 144–180 m.a.s.l. The north-west half is dominated by deciduous forest mainly consisting of oak (*Quercus serrata* Murray), Japanese snowbell (*Styrax japonica* L.), and Japanese loose-flowered Hornbeam (*Carpinus laxiflora* (Sieb. et Zucc.) Blume). Japanese cedar (*Cryptomeria japonica* (L.f.) D. Don) was planted at the south-east part (Fig. 1b). Steep slopes are found at the center and southern part of the plot, while the area on the western side is quite flat (Fig. 1a). The topological characteristics of the sub-plots were categorized into five groups according to the slope angle ( $\theta$ ), slope direction ( $d$ ), elevation ( $e$ ), and a vegetation map (Fig. 1c). The categorization was conducted as follows: valley sub-plots (Valley)—sub-plots with  $e < 160$  m and  $\theta < 12^\circ$ ; hill top sub-plots (Top)—sub-plots with  $e > 170$  m and  $\theta > 20^\circ$ ; north-slope coniferous trees dominating sub-plots (North-Conifer)—sub-plots with a proportion of coniferous trees more than 35%,  $d < 90^\circ$  or  $d > 270^\circ$ , except plots already categorized as Valley or Top; north-slope deciduous trees dominating sub-plots (North-Deciduous)—sub-plots with a proportion of coniferous trees less than 35%,  $d < 90^\circ$  or  $d > 270^\circ$ , except plots already categorized as Valley or Top; and south-slope sub-plots with only deciduous forest (South-Slope)—sub-plots with  $90^\circ < d < 270^\circ$ , except plots already categorized as Valley or Top. The sub-plot numbers categorized as Valley, North-Conifer, North-Deciduous, South-Slope, and Top are 11, 14, 14, 39, and 22, respectively. The soil in the sampling site is a dystric cambisol (FAO/UNESCO 1990) derived from Tama loam, a volcanic ash layer, with a very low bulk density around 0.7 g cm<sup>-3</sup>, except at the Valley, which comprised compacted fluvisols (FAO/UNESCO 1990).

### Gas and Soil Sampling and Measurement of Environmental Factors

Carbon dioxide, CH<sub>4</sub>, and N<sub>2</sub>O fluxes from soil surfaces were measured during the full vegetative period on the 25th of July, 2008 and the litter fall period on the 5th of December, 2008. No rain was recorded 7 days before both sampling days. The average temperature was 29.0 °C on the 25th of July and 11.6 °C on the 5th of December (Japan Meteorological Agency 2010). The monthly average temperature in July and December in 2008 was 26.1 and 7.1 °C, respectively. Soil moisture content monitored continuously with a PR2/4 profile probe (DIK-350D) fluctuated from 44 to 82% at a mixed forest site on a slope position near the study area. The gas was sampled using the closed-chamber method according to Rolston (1986).



**Fig. 1** General characteristics of the study site, JaLTERplot: **a** elevation, **b** vegetation distribution, and **c** slope direction and topological groups. Topological groups of the sub-plot are as follows: *Valley* Valley, *North-Conifer* North slope coniferous trees, *North-Deciduous* North slope deciduous trees, *South-Slope* South slope deciduous trees, *Top* Top of the hill. A–B and 1–10 are the IDs of sub-plots

Hundred chamber bases were inserted into the soil to a depth of 5 cm on the 11th of July, 2008 at the center of the sub-plots. The chamber had a diameter of 20.2 cm and height of 15.0 cm, and thus a volume of 9609 cm<sup>3</sup>. All gas sampling was conducted during 13:00 and 15:00 by measuring 20 places at once. A previous experiment showed that GHG fluxes from soil could be sufficiently detected within 20 min (Bekku et al. 1995). Thus, gas samples were taken 1, 7, 15, and 21 min after the closure of the chamber.

During and immediately after the gas sampling, air and soil temperature were measured at a depth of 5 cm from a position surrounding the chamber and inside the chamber. After the gas sampling, a composite soil sample from a depth of 0–5 cm was collected from a position surrounding each chamber in July, and from inside the chamber in December. The litter layer inside the chamber was also sampled in December.

**Gas Analyses**

The concentrations of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O in the sample vials were analyzed no later than 10 days after sampling using an automated analysis system (Sudo 2006). This system consisted of two gas chromatographs (GC-14B, Shimadzu, Kyoto, Japan), one of which had a thermal conductivity detector and a flame ionization detector (FID), and the other possessed an electron capture detector (ECD). Calibration of the GC system was conducted using more than two concentrations of each standard gas. The concentrations of gas from each chamber were plotted against the sampling time and all fitted a linear change in gas concentration ( $R^2 > 0.90$  for CH<sub>4</sub> and N<sub>2</sub>O;  $R^2 > 0.99$  for CO<sub>2</sub>).

**Analyses of Soil and the Litter Layer**

Soil water content was determined by comparing the fresh weight and dry weight (after drying at 105 °C for 2 days).

Soil samples were then sieved (2-mm mesh) and part of the fresh soil was used for KCl extraction, while the remaining portion was air-dried for total carbon (TC) and total nitrogen (TN) analyses (as described later). The water content of the litter layer was determined by the difference between the weight of the fresh soil and the weight after oven drying at 80 °C for 2 days. All litter layer samples were then sieved (8-mm mesh) and measured for dry weight. The TC and TN of soil and the litter layer were measured using an element analyzer (lash EA112, ThermoQuest, Italy).

Soil pH (H<sub>2</sub>O) was determined from a solution comprising 10 g of fresh soil and 25 ml of deionized water as measured by a glass electrode (Handy-type pH meter D-51S, Horiba). Five-gram samples of fresh soil (< 2 mm) were extracted with 50 ml of a 2 M KCl solution for 1 h in a 100-ml bottle. Nitrate was analyzed using the copper–cadmium reduction method, and inorganic NH<sub>4</sub><sup>+</sup>-N was analyzed using the indophenol blue method in a continuous-flow analyzer (TRAACS, BranþLuebbe, Norderstedt, Germany).

**Statistical Analysis**

A minimum sample of 100 points is required to compute valuable semivariograms (Webster and Oliver 1992; Lark 2000). The plotting of a semivariogram is a method that can be used to express the spatial dependency of soil parameters (Yanai and Kosaki 2000). In the semivariogram graph, the semivariance is plotted in relation to the distance between measurement points *h*. The semivariance is the mean square difference of the variance at a given distance *h*. If the soil parameters are spatially dependent, the semivariance reaches a maximum known as a sill. The distance *h* that reaches the sill is known as the range. If the lag, which is the distance among the sample numbers used

for the analysis, equals 0, the semivariance should become 0 because the value should refer to the same data. However, due to unaccounted variability such as variability within the sampling intervals or measurement error, the semivariance attains a positive value known as the nugget. Long ranges indicate that the soil parameters are spatially dependent for a wide area. The relation between the sill and nugget is expressed by the  $Q$  value, which is given in the following equation:

$$Q \text{ value} = (S - N)/S, \quad (1)$$

where  $S$  and  $N$  represent the sill and nugget, respectively. If the  $Q$  value of a variable is close to 1, this indicates that the spatial dependency of the variable is high. Soil parameters with a  $Q$  value larger than 0.5 are considered spatially dependent. If the  $Q$  value is smaller than 0.5, this means that the unaccounted variability is larger than the spatial variability.

A test for normality of the sample distribution was carried out using the Kolmogorov–Smirnov test (SigmaStat 3.1, Systat Software Inc.). Variograms were created on the basis of the distribution pattern, which was normal, log-normal, or no-normality. The spatial distributions of greenhouse gas fluxes and soil properties were determined using the Add-in Geostatistical Analyst of ArcGIS 9.1 (ESRI Inc.). Log-normally distributed data were log-transformed for the calculations. If the data were spatially dependent, a semivariogram with the smallest residual sum of squares was created by ordinary kriging to obtain a prediction map. If the  $Q$  value was below 0.5, the inverse distance weighting method was used to obtain a prediction map. The spatial dependency was determined from the  $Q$  value, range, and regression coefficient (Konda et al. 2008).

## RESULTS

### Soil and Litter Layer Properties

All  $Q$  values of soil and litter properties, with the exception of the water content of the litter layer, were high and ranged from 0.51 to 0.79, which reflect a strong spatial dependence (Table 1). The ranges of all soil parameters, including the dry weight and TN content of the litter layer, were similar and ranged from 49.5 to 59.3 m, while the range of the TC and CN ratio of the litter layer was shorter and equaled 19.7 m. The regression coefficient of all soil parameters was high and ranged from 0.54 to 0.68, confirming their strong spatial dependence, while parameters of the litter layer were lower and ranged from 0.21 to 0.59. The spatial dependency was weakest for total C, which had the lowest regression coefficient among litter layer

properties. The water content of the litter layer did not exhibit any spatial dependency.

A significant topological difference was found after soil parameters were divided into topological groups as defined in Fig. 1c. Total C and TN were highest in Top and lowest in North-Conifer and North-Deciduous groups (Fig. 2a, c). The pH was highest in Valley, followed by Top (Fig. 2g). While TC, TN and pH tended to increase from North-Conifer to North-Deciduous, South-Slope, Top and Valley, the CN ratio was significantly higher in North-Conifer compared to the other topological groups, and there was no significant difference among the other categories (Fig. 2e). There was no large topological difference for the litter layer regarding TC concentration, while TN was significantly highest in Top and lowest in North-Conifer (Fig. 2b, d). The CN ratio of North-Conifer and Valley was significantly higher than that of the other topological groups (Fig. 2f). The amount of dry weight was highest in Top and lowest in Valley (Fig. 2h).

### Seasonal Changes in Soil Conditions

High  $Q$  values of more than 0.75 were found for soil moisture and soil  $\text{NH}_4^+$ -N concentration at both sampling times (Table 1). Soil temperature showed a higher spatial dependency ( $Q$  value: 1.00,  $r^2$ : 0.85) in December than in July ( $Q$  value: 0.53,  $r^2$ : 0.45). Nitrate nitrogen showed weak spatial dependency in July ( $Q$  value: 0.50,  $r^2$ : 0.35) and no spatial dependency in December. The ranges were wider in December than in July for soil temperature and  $\text{NH}_4^+$ -N, while it was lower for soil moisture in December compared to July. Overall,  $Q$  values and regression coefficients were higher in December than in July.

Soil moisture was significantly ( $P < 0.001$ ) lower in July than in December, and soil temperature was significantly ( $P < 0.001$ ) higher in July than in December. The differences among topological groups were the same for soil moisture for both sampling seasons, and the Valley group showed significantly higher values than those of the other topological groups (Fig. 3a). Soil temperature of the South-Slope and Top groups was significantly lower than that of the other topological groups in July; however, South-Slope and Top showed a significantly higher soil temperature in December (Fig. 3b). Nitrate nitrogen was significantly ( $P < 0.001$ ) higher in July than in December, but  $\text{NH}_4^+$ -N showed no significant difference between the sampling times (Fig. 3c, d). The difference among topological groups was smaller in December compared to July. The trend was similar when comparing July and December for  $\text{NO}_3^-$ -N, which showed a higher value for North-Conifer compared to North-Deciduous. However, the trend differed for  $\text{NH}_4^+$ -N, which exhibited lower values in July for

**Table 1** Geostatistical values for CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O in July and December

	Property	Unit	Nugget	Sill	Range	Q value	r <sup>2</sup>	Model
Basic soil properties at 0–5 cm soil	Total C	(Log mg C g <sup>-1</sup> )	0.007	0.020	59.3	0.66	0.61	E
	Total N	(Log mg N g <sup>-1</sup> )	0.005	0.021	59.3	0.79	0.68	E
	C/N ratio	Ratio	0.40	1.70	59.3	0.76	0.66	E
	pH (H <sub>2</sub> O)		0.049	0.232	49.5	0.79	0.54	E
Litter layer	Total C	(mg C g <sup>-1</sup> )	1601	3885	19.7	0.59	0.21	S
	Total N	(mg N g <sup>-1</sup> )	4.3	8.7	59.3	0.51	0.45	E
	C/N ratio	Ratio	5.1	21.6	19.7	0.76	0.59	S
	Dry weight	(Log g m <sup>-2</sup> )	0.023	0.068	59.3	0.66	0.57	E
	Water content	(%)	–	–	–	0.19	–	IDW
Soil condition	July							
	Soil moisture	(Log %)	0.005	0.018	30.2	0.75	0.56	S
	Soil temperature	(°C)	0.18	0.39	42.9	0.53	0.45	S
	NO <sub>3</sub> <sup>-</sup> -N	(Log mg N g <sup>-1</sup> )	0.019	0.038	24.7	0.50	0.35	S
	NH <sub>4</sub> <sup>+</sup> -N	(mg N g <sup>-1</sup> )	1.1 × 10 <sup>-5</sup>	9.7 × 10 <sup>-5</sup>	26.1	0.88	0.50	E
	December							
	Soil moisture	(Log %)	0.000	0.018	24.4	1.00	0.54	E
	Soil temperature	(°C)	0.00	0.66	59.3	1.00	0.85	E
NO <sub>3</sub> <sup>-</sup> -N	(mg N g <sup>-1</sup> )	–	–	–	0.23	–	IDW	
NH <sub>4</sub> <sup>+</sup> -N	(Log mg N g <sup>-1</sup> )	0.0	2.8	39.0	1.00	0.81	E	
Gas fluxes	July							
	CO <sub>2</sub> flux	(mg C m <sup>-2</sup> h <sup>-1</sup> )	–	–	–	0.06	–	IDW
	CH <sub>4</sub> flux	(μg cm <sup>-2</sup> h <sup>-1</sup> )	369	1126	62.2	0.67	0.38	E
	N <sub>2</sub> O flux	(μg N m <sup>-2</sup> h <sup>-1</sup> )	–	–	–	0.09	–	IDW
	December							
	CO <sub>2</sub> flux	(mg C m <sup>-2</sup> h <sup>-1</sup> )	–	–	–	0.31	–	IDW
CH <sub>4</sub> flux	(μg cm <sup>-2</sup> h <sup>-1</sup> )	48	502	72.0	0.90	0.48	S	
N <sub>2</sub> O flux	(μg N m <sup>-2</sup> h <sup>-1</sup> )	–	–	–	0.01	–	IDW	

S spherical, E exponential, IDW Inverse distance weighted

North-Conifer and Valley compared to the other groups, and higher values in December.

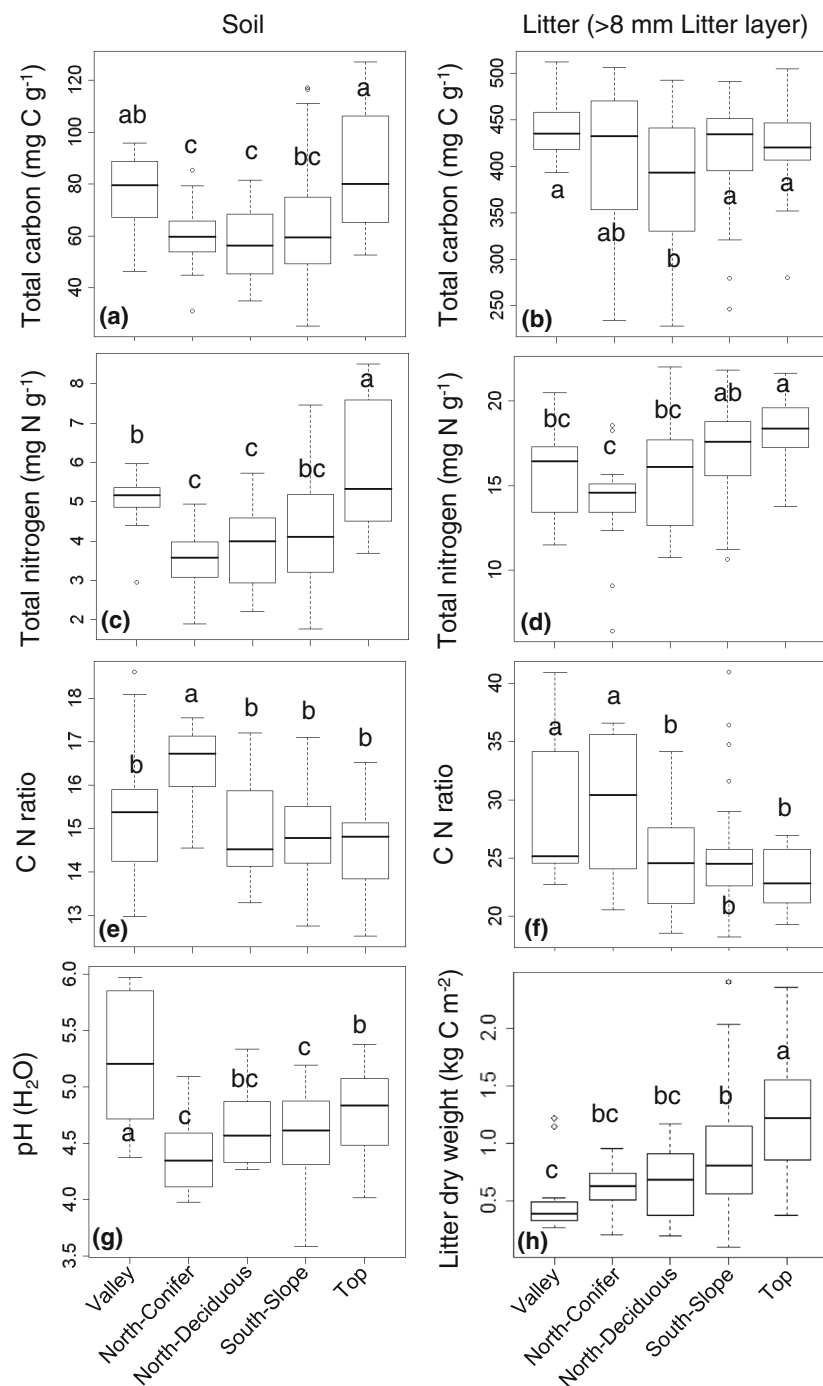
**Greenhouse Gas Fluxes**

Q values for CO<sub>2</sub> and N<sub>2</sub>O fluxes in both seasons were less than 0.50 and showed no spatial dependency (Table 1). On the other hand, the Q value of CH<sub>4</sub> was high and equaled 0.67 in July and 0.90 in December, indicating strong spatial dependency even though the regression coefficients were lower than those of soil parameters. The range of CH<sub>4</sub> in both July and December was wide and equaled 62 m in July and 72 m in December, showing a spatial dependency over a long distance.

All three gas fluxes differed significantly between July and December, and the emissions of CO<sub>2</sub> and N<sub>2</sub>O, as well as CH<sub>4</sub> uptake, were higher in July than in December (Fig. 4). However, the topology showed a different influence on each gas. The CO<sub>2</sub> flux showed no topological difference in July, but exhibited a significantly lower value in North-Conifer compared to the other topological groups

in December (Fig. 4a). The trend for the CH<sub>4</sub> flux was the same for both July and December, and showed a declining tendency from North-Conifer, North-Deciduous, South-Slope, and Top, while Valley showed a significantly higher value at both sampling times (Fig. 4b). The N<sub>2</sub>O flux showed a significantly higher value for the Valley group in July, but no topological difference was found in December (Fig. 4c).

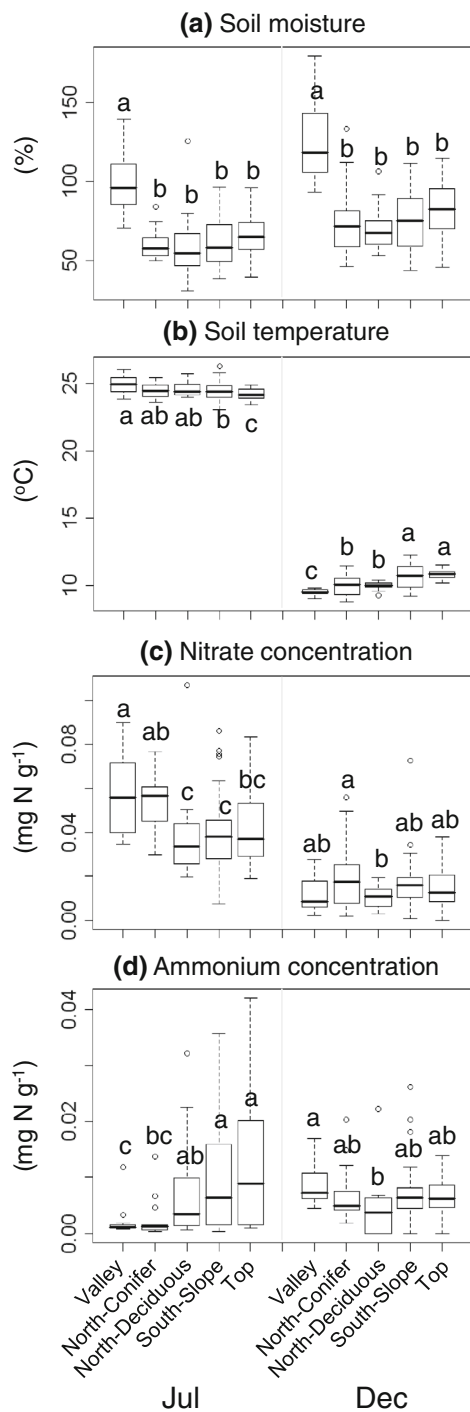
The three greenhouse gases under investigation showed distinct characteristic distributions for the two dates (Fig. 5). The CO<sub>2</sub> flux showed an overall tendency to decline from the north to the south (Fig. 5a, d). This tendency was much clearer in December compared to July, although the flux was much higher in July than in December. The CH<sub>4</sub> flux was clearly influenced by the topology at both sampling times (Fig. 5b, e). The two valley areas showed the highest fluxes, and exhibited CH<sub>4</sub> emission points of 6 and 1 for July and December, respectively. The lowest fluxes were found in the South-Slope and North-Deciduous groups in July and the South-Slope and Top groups in December. The N<sub>2</sub>O flux



**Fig. 2** Boxplots of soil and litter (>8 mm litter layer) properties for the five topological groups: **a, b** total carbon (TC), **c, d** total nitrogen (TN), **e, f** CN ratio of soil and litter layer, respectively, **g** pH of soil, and **h** dry weight of litter layer. The line in the box indicates the median (50% percentile). The upper and lower borders of each box mark the means of the 25 and 75% percentiles, respectively. The whiskers (vertical dashed lines above and below the box) show the largest and smallest observed values except for existing outliers. The outliers (open circles) represent the values those are larger (or smaller) than 1.5 times the box length from the 75% percentile (or 25% percentile). Superscript letters denote statistical similarities and differences between sample groups within each

showed high emission in the two valley areas in July and reached  $298 \mu\text{g N m}^{-2} \text{h}^{-1}$  at point I4 (Fig. 5c). Low fluxes were found in the slope and hill-top areas, while the distribution of low fluxes was much more random

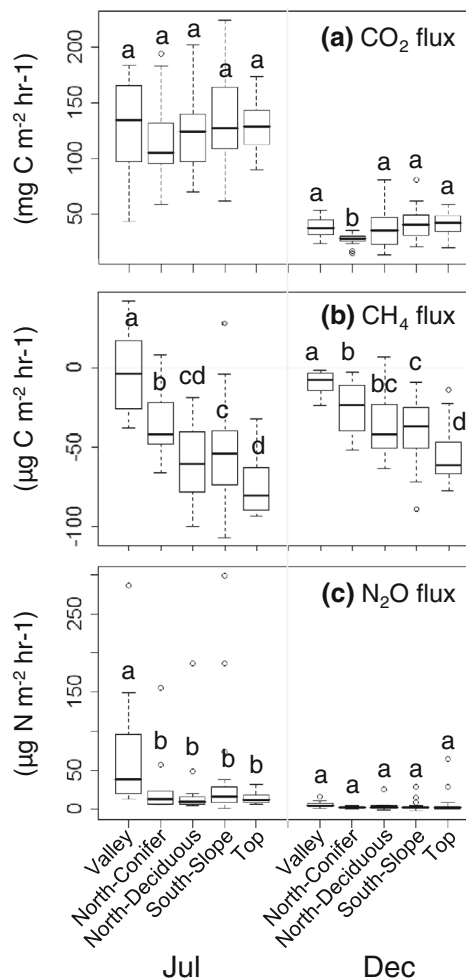
than that of  $\text{CO}_2$  or  $\text{CH}_4$  fluxes during the same period. The distribution of the  $\text{N}_2\text{O}$  flux in December differed completely from that in July (Fig. 5f). Only a few points showed high fluxes.



**Fig. 3** Boxplots of soil properties for the five topological groups in July and December: **a** soil temperature, **b** soil moisture, **c** soil NO<sub>3</sub>-N, and **d** NH<sub>4</sub><sup>+</sup>-N concentration. The description of boxplot is given in Fig. 2

**Simple Regression Analysis Among Investigated Parameters**

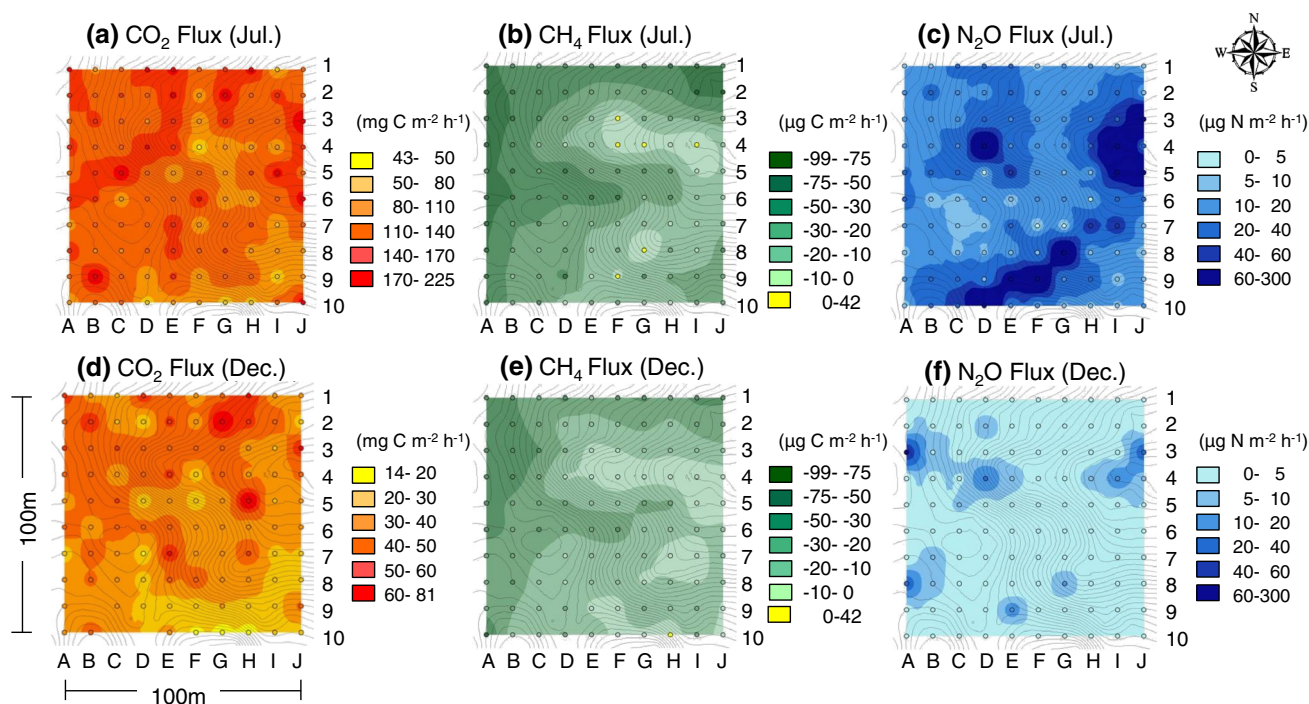
Carbon dioxide fluxes showed a high correlation ( $r = 0.50$ ,  $P < 0.01$ ) between the two sampling times in July and



**Fig. 4** Boxplots of greenhouse gas fluxes for the five topological groups in July and December: **a** CO<sub>2</sub>, **b** CH<sub>4</sub>, and **c** N<sub>2</sub>O fluxes. The description of boxplot is given in Fig. 2

December. In both seasons, CO<sub>2</sub> fluxes showed a significant positive correlation ( $P < 0.05$ ) with soil NH<sub>4</sub><sup>+</sup>-N ( $r = 0.36$  and  $r = 0.33$ , respectively), as well as dry weight ( $r = 0.25$  and  $r = 0.33$ , respectively) and TN of the litter layer ( $r = 0.27$  and  $r = 0.30$ , respectively). In July, the CO<sub>2</sub> flux showed a significant positive correlation ( $P < 0.05$ ) with the N<sub>2</sub>O flux ( $r = 0.41$ ), soil moisture ( $r = 0.25$ ), TC ( $r = 0.26$ ) and TN ( $r = 0.30$ ) of soil, and the water content ( $r = 0.30$ ) of the litter layer. In December, the CO<sub>2</sub> flux showed a significant negative correlation ( $P < 0.05$ ) with the CH<sub>4</sub> flux ( $r = -0.37$ ) and CN ratio of soil ( $r = -0.24$ ).

The CH<sub>4</sub> fluxes showed a highly significant positive correlation ( $P < 0.001$ ) in July and December ( $r = 0.85$ ). The CH<sub>4</sub> fluxes in both seasons showed a significant negative correlation ( $P < 0.05$ ) with soil NH<sub>4</sub><sup>+</sup>-N in July ( $r = -0.29$  and  $r = -0.25$ , respectively), soil temperature in December ( $r = -0.48$  and  $r = -0.49$ , respectively), TC ( $r = -0.30$  and  $r = -0.38$ , respectively) and TN



**Fig. 5** Spatial distribution of greenhouse gases in July and December for CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O, respectively

( $r = -0.36$  and  $r = -0.42$ , respectively) of soil, dry weight ( $r = -0.47$  and  $r = -0.44$ , respectively), and TN ( $r = -0.27$  and  $r = -0.27$ , respectively) of the litter layer. In July, the CH<sub>4</sub> flux showed a significant positive correlation ( $P < 0.05$ ) with the N<sub>2</sub>O flux ( $r = 0.36$ ), soil moisture ( $r = 0.27$ ), and the CN ratio of the litter layer ( $r = 0.26$ ). No additional correlations were found for soil parameters in December.

Nitrous oxide fluxes also showed a significant positive correlation in July and December, but it was weaker than that of CO<sub>2</sub> and CH<sub>4</sub> ( $r = 0.35$ ,  $P < 0.05$ ). In both seasons, a significant positive correlation ( $P < 0.05$  and  $P < 0.01$ ) was found with soil moisture ( $r = 0.54$  and  $r = 0.29$ , respectively) and soil pH ( $r = 0.30$  and  $r = 0.28$ , respectively). There was a significant positive correlation ( $P < 0.05$ ) with the water content of the litter layer in July ( $r = 0.38$ ), and a significant negative correlation ( $P < 0.05$ ) with soil NH<sub>4</sub><sup>+</sup>-N in December ( $r = -0.36$ ).

Among the significant correlations of soil and litter layer parameters, remarkable was the high correlation ( $P < 0.01$ ) between soil moisture and soil NO<sub>3</sub><sup>-</sup>-N in July ( $r = 0.56$ ), and pH ( $r = 0.56$ ), TC ( $r = 0.57$ ) and TN ( $r = 0.58$ ). Soil moisture was highly positively correlated ( $P < 0.001$ ) between the two sampling times ( $r = 0.77$ ). As expected, TC and TN showed a high positive correlation for soil and litter ( $r = 0.97$ ,  $P < 0.001$  and  $r = 0.24$ ,  $P < 0.05$ , respectively).

## DISCUSSION

### Topological Differences in CO<sub>2</sub> Flux

In this study, the CO<sub>2</sub> flux from the soil involves both heterotrophic respiration and root respiration. The contribution of heterotrophic respiration to the total CO<sub>2</sub> flux from the soil can range from 27 to 53% in forest soil (Ekblad and Högberg 2001). Both heterotrophic and root respirations are influenced by soil moisture and soil temperature. Soil and litter CN ratios are also important factors that influence heterotrophic respiration. Moderate moisture, higher temperature, and a low CN ratio usually increase the CO<sub>2</sub> flux, while water-saturated or overly dry soil, low temperature, and a high CN ratio decrease the CO<sub>2</sub> flux. Summer will therefore show a higher CO<sub>2</sub> flux compared to that of winter, which was found in this study.

Significant positive correlations between CO<sub>2</sub> fluxes and litter layer parameters indicate that decomposition of organic matter is determining CO<sub>2</sub> emission. Soil moisture and temperature, which determine microbial activity, showed significant differences among the topological groups in July and December (Fig. 3a, b). However, there was no significant difference for CO<sub>2</sub> flux among topological groups in July (Fig. 5a). The flux of the North-Conifer sub-plot groups tended to be low, and topology showed no significant influence on CO<sub>2</sub> emission in July. In December, North-Conifer showed a significantly lower



CO<sub>2</sub> flux compared to the other topological groups. The northern slope receives less sunshine during winter due to a lower angle of the sun, which results in a lower soil temperature (Fig. 3b). Additionally, needles on coniferous trees do not fall in winter and keep the soil under a shadow throughout the year, whereas deciduous trees lose their leaves in winter and thereby allow sunshine to reach the ground and provide the soil with organic matter. The CN ratio was significantly higher at North-Conifer (Fig. 2e, f) due to the litter provided by coniferous species, which led to the negative correlation between the CO<sub>2</sub> flux and CN ratio in December. Increased N deposition is known to enhance C cycling in a forest (Hutyara et al. 2011). High CO<sub>2</sub> emission due to the deposition masked the topological differences in July, and only a small difference was found in December.

### Topological Differences in CH<sub>4</sub> Flux

The most pronounced characteristic of our site is the high N deposition (Kimura et al. 2009). Addition of NH<sub>4</sub><sup>+</sup>-N to the soil led to reduced CH<sub>4</sub> uptake by the soil because ammonium is more favorable for methanotrophs than CH<sub>4</sub> (Gulledge et al. 1997). The CH<sub>4</sub> uptake fluxes recorded in this study (Fig. 4b) are smaller than those obtained from a similar volcanic forest soil without slopes at Tsukuba (about 130 μg C m<sup>-2</sup> h<sup>-1</sup>, Yonemura et al. 2000). However, the range of flux values in our study (3–175 μg C m<sup>-2</sup> h<sup>-1</sup>) is comparable to that recorded for other Japanese soils (Morishita et al. 2007). The frequency of measurements are not the same for the compared studies; however, these results show that a high amount of N deposition did not completely inhibit CH<sub>4</sub> oxidation and the topological differences had a significant influence on the CH<sub>4</sub> flux.

Methane is emitted from anaerobic soils by methanogens and is oxidized in aerobic soils by methanotrophs (Conrad 2007). Thus, CH<sub>4</sub> is emitted from soil under high-moisture conditions and is absorbed by soils under low-moisture conditions. In our study, we found CH<sub>4</sub> emission in the Valley group and a kind of hollow (water-pass) part of the North-Conifer group (Fig. 5b, e). The overall CH<sub>4</sub> uptake flux distribution reflects the distribution of soil moisture, and CH<sub>4</sub> uptake (negative flux) was larger under low soil-moisture conditions. The low soil moisture observed in South-Slope and Top groups favors greater soil gas diffusivity and increased CH<sub>4</sub> uptake activity of methanotrophs, as noted by many previous studies (Dörr et al. 1993; Konda et al. 2008).

The distribution of soil moisture was similar in July and December (Fig. 3a) and the CH<sub>4</sub> flux distributions therefore did not differ greatly (Fig. 4b), as evidenced by the high positive correlation ( $P < 0.001$ ). Soil parameters correlated

with CH<sub>4</sub> fluxes are related to the soil redox condition, although a direct correlation between CH<sub>4</sub> and soil moisture was only found in July. The moisture content distribution was narrower in December than in July, and some high emissions were also found at the Top position such as H10. Methane uptake fluxes in July were larger than those in December (Fig. 4b). This is attributable to higher soil temperature and lower soil moisture in July, leading to greater soil gas diffusivity and increased activity of methanotrophs. Similarly, the magnitude of CH<sub>4</sub> emission fluxes and the number of CH<sub>4</sub>-emitted points are greater in July than December. This is due to the larger temperature dependence of CH<sub>4</sub> emission by methanogens than that of CH<sub>4</sub> uptake by methanotrophs (Whalen and Reeburgh 1996).

The CH<sub>4</sub> flux showed high spatial dependency among all soil properties (Table 1). Our results showed that the nugget is much smaller than the sill. If the nugget is bigger than the sill, the unaccounted error is larger than the analyzed spatial variance and indicates the limited significance of the analyzed spatial dependency range (Konda et al. 2008). The strong spatial dependency of the CH<sub>4</sub> flux is also confirmed by the high  $Q$  value and regression coefficient. The high correlation between CH<sub>4</sub> fluxes and soil moisture might be the reason for this strong dependency. The spatial dependency of the CH<sub>4</sub> flux obtained in this study is clearer than that demonstrated for tropical soils (Konda et al. 2008; Fang et al. 2009). One reason for this clarity is that our soil showed greater CH<sub>4</sub> uptake than that recorded for tropical soils, even under high N deposition. Another reason is that termites usually contribute to the CH<sub>4</sub> emission in tropical soils, which may mask the influence of topology.

### Topological Differences in N<sub>2</sub>O Flux

The N<sub>2</sub>O fluxes of the present study are high compared to values recorded elsewhere in Japan (Morishita et al. 2007). The high N deposition in this site (Kimura et al. 2009) stimulated the N<sub>2</sub>O emission (Hatano et al. 2007). In addition, gas sampling in a forest is usually conducted at representative points (Sakata et al. 2004), whereas various points with different topology were measured in our study. The complex topology that included hot spots such as those in the Valley topological group increased the average flux. Possible hot spots of N<sub>2</sub>O emission must be taken into account more often when evaluating N<sub>2</sub>O emission from a forest soil.

As in previous studies (Morishita et al. 2007; Nishina et al. 2009a), the present study showed high N<sub>2</sub>O fluxes in July and low fluxes in December (Fig. 4c). Seasonal variability in Japan originates from microbial activity involved in N<sub>2</sub>O formation under conditions of high temperature and

humidity in July (Morishita et al. 2007). The difference in average soil temperature between July and December was 14.0 °C (Fig. 3b), which led to 85% lower N<sub>2</sub>O fluxes in December compared to July.

The present study showed a high N<sub>2</sub>O flux for the Valley group and surrounding area compared to the other topological groups in both July and December (Fig. 4c). This trend has also been observed in other studies (Osaka et al. 2006; Fang et al. 2009; Nishina et al. 2009b) and can be explained by stimulation of N<sub>2</sub>O production through nitrification and denitrification under increased soil moisture (Papen et al. 2001). On the other hand, a high N<sub>2</sub>O flux was observed at A3 and A8 in December (Fig. 5f). Hot spots of N<sub>2</sub>O emission were also reported by Nishina et al. (2009b). Measurements in their study were made at 1-m intervals; however, since the nugget value of 1.17 was considerably higher than the sill (2.45), they concluded that even an intensive measurement method involving 1-m intervals was not sufficient to determine spatial factors influencing N<sub>2</sub>O emission. At a small scale, the existence of anaerobic micro-sites is thought to be the cause for the presence of hot spots (Nishina et al. 2009b). Anaerobic micro-sites are reported as the reason for denitrification by many studies (Meijide et al. 2010). The topological group Top showed accumulation of litter (Fig. 2h) and higher soil moisture (Fig. 3a), which led to the formation of anaerobic micro-sites such as A3 and A8. The high potential for N<sub>2</sub>O emission due to N deposition and radiation onto the forest floor during winter led to N<sub>2</sub>O emission in December. The hot spots of N<sub>2</sub>O emission thus differed between July and December. While CO<sub>2</sub> and CH<sub>4</sub> showed an apparently constant spatial dependency in this study, N<sub>2</sub>O hotspots are reported to vary highly over time (Röver et al. 1999), and more intensive sampling intervals might identify other points as hot spots.

## CONCLUSION

It can be concluded that CH<sub>4</sub> and N<sub>2</sub>O emissions from an urban forest were highly variable throughout the landscape, while CO<sub>2</sub> emission was more evenly distributed. Slope direction and position had a high influence on soil characteristics, and combined with vegetation type to yield site-specific GHG emissions. High N deposition enhanced the CO<sub>2</sub> emission and masked the topological differences, while no limitation in available N due to high N deposition led to high N<sub>2</sub>O fluxes that emphasize the controlling factors related to spatial variability such as water conditions and organic C supply. The spatial dependency of CH<sub>4</sub> fluxes was stronger than that of CO<sub>2</sub> or N<sub>2</sub>O, with a constant high emission in the valley, thus confirming the

importance of soil water status. To estimate the GHG emission from natural ecosystem more accurately, it is essential to take the topological variability into account.

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## AUTHOR BIOGRAPHIES

**Sonoko Dorothea Bellingrath-Kimura** (✉) has a Ph.D. in agriculture from Hokkaido University, Japan. She is an associate professor at the Graduate School of Agriculture at Tokyo University of Agriculture and Technology (TUAT), in Japan. Her research covers a wide area that relate on nutrient cycling in agro- and forestry-ecosystems. She summarizes her work in the Eco-balance analysis. *Address:* Department of International Environmental and Agricultural Science, Graduate School of Agriculture, Tokyo University of Agriculture and Technology, Saiwaicho 3-5-8, Fuchu, Tokyo 183-8509, Japan. e-mail: skimura@cc.tuat.ac.jp

**Ayaka Wenhong Kishimoto-Mo** has a Ph.D. in agriculture from Gifu University, Japan. She is a senior researcher at the National Institute for Agro-Environmental Sciences (NIAES), Japan. Her research covers carbon dynamics in terrestrial ecosystems (forest, grassland, and tundra) and greenhouse gases mitigation in upland cropland. She is an expertise on standardization of chamber method for long-term monitoring soil CO<sub>2</sub> efflux and other greenhouse gases. *Address:* Carbon and Nutrient Cycles Division, National Institute for Agro-Environmental Science, Tsukuba 305-8604, Japan. e-mail: mow@affrc.go.jp

**Noriko Oura** has a Ph.D. in science from Shinshu University, Japan. She is a senior researcher at the National Institute for Agro-Environmental Sciences (NIAES), Japan. Her research covers nitrogen dynamics in agro- and forestry-ecosystems and its greenhouse gases (N<sub>2</sub>O). She investigates organic matter application as greenhouse gases mitigation measure in cropland. *Address:* Carbon and Nutrient Cycles Division, National Institute for Agro-Environmental Science, Tsukuba 305-8604, Japan. e-mail: nori@affrc.go.jp

**Seiko Sekikawa** has a Ph.D. in agriculture from Gifu University, Japan. He is a professor at the Department of Agriculture at the Tamagawa University, in Japan. His research covers a wide area that relate on carbon dynamics of plant and soil processes in agro-, grassland-, and secondary forest ecosystems. *Address:* College of Agriculture, Tamagawa University, Machida 194-8610, Japan. e-mail: sekisei@agr.tamagawa.ac.jp

**Seichiro Yonemura** has a Ph.D. in science from University of Tokyo, Japan. He is a senior researcher at National Institute for Agro-Environmental Sciences (NIAES), in Japan. His research covers a wide area that relate on gas exchange between the atmosphere and ecosystems.

*Address:* Agro-Meteorology Division, National Institute for Agro-Environmental Science, Tsukuba 305-8604, Japan.  
e-mail: yone@affrc.go.jp

**Shigeto Sudo** has a Ph.D. in science from Tokyo Institute of Technology, Japan. He is a senior researcher at the Carbon and Nutrient Cycles Division, National Institute for Agro-Environmental Science (NIAES). He develops sophisticated and simplified monitoring systems to establish mitigation techniques of greenhouse gas from farmland soils.

*Address:* Carbon and Nutrient Cycles Division, National Institute for Agro-Environmental Science, Tsukuba 305-8604, Japan.  
e-mail: ssudo@affrc.go.jp

**Atsushi Hayakawa** has a Ph.D. in agriculture from Hokkaido University, Japan. He is an associate professor at Department of Biological Environment at Akita Prefectural University, in Japan. His research focuses on nutrient biogeochemical cycles such as nitrogen and phosphorus in watershed scale.

*Address:* Faculty of Bioresource Sciences, Akita Prefectural University, Akita 010-0195, Japan.  
e-mail: hayakawa@akita-pu.ac.jp

**Kazunori Minamikawa** has a D.Agr. from the University of Tsukuba, Japan. He is now a researcher at the National Institute for Agro-Environmental Sciences in Japan. His research subject is measurement and

modeling of greenhouse gas emissions from agro-ecosystems.

*Address:* Carbon and Nutrient Cycles Division, National Institute for Agro-Environmental Science, Tsukuba 305-8604, Japan.  
e-mail: minakazu@affrc.go.jp

**Yusuke Takata** has a Ph.D. in agriculture from Kyoto University, Japan. He is a researcher at National Institute for Agro-Environmental Sciences (NIAES) in Japan. His research field is a spatial pattern analysis of soil properties in agro-ecosystems.

*Address:* Natural Resources Inventory Center, National Institute for Agro-Environmental Science, Tsukuba 305-8604, Japan.  
e-mail: takatay@affrc.go.jp

**Hiroshi Hara** has a Ph.D. degree in physical chemistry from Tokyo Institute of Technology, Japan. He is now a professor emeritus at Tokyo University of Agriculture and Technology, and continues his work on precipitation chemistry based on national, regional, and global monitorings in which he has been deeply involved ever since. Asian history of atmospheric chemistry is also his research focus to summarize his career in a larger academic sense.

*Address:* Department of Agriculture, Tokyo University of Agriculture and Technology, Fuchu 183-8509, Tokyo, Japan.  
e-mail: harahrs@cc.tuat.ac.jp