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Nitrous Oxide (N_2O) Emissions from a Japanese Lowland Soil Cropped to Onion: I. Spatial and Temporal Variability of Fluxes

[1]Nsalambi V. Nkongolo, [2]Kanta Kuramochi and [2]Hatano Ryusuke
[1]Department of Agriculture and Environmental Sciences,
College of Agricultural and Natural Sciences, Lincoln University of Missouri,
Jefferson City, MO 65102-0029, USA
[2]Field Science Center for the Northern Biosphere, Laboratory of Soil Science,
Graduate School of Agriculture, Hokkaido University, Sapporo, Hokkaido 060-8589, Japan

Abstract: Field studies were conducted to assess the spatial and temporal variability of nitrous oxide (N_2O) emissions in an agricultural field cropped to onion in Mikassa, northern Hokkaido (Japan). N_2O emissions measurements were conducted in 100 by 100 m and 60 by 60 m grids in 1999 and 2000, respectively with samples taken at 10 m spacing. Air samples for N_2O determinations were collected using the closed-chamber technique. The chambers were circular with steel frames. The top of each chamber had a gas sampling tube and a bag to control air pressure inside. The height and diameter of the chamber were 0.35 and 0.30 m, respectively. Air samples were stored in vial bottles for analysis with a gas chromatograph with electron capture detector within 24 h after sampling. GS+ 7.0 geostatistical software and statistix 8.0 were used for data analysis. Results showed that N_2O emissions were highest in 1999 as compared to 2000. N_2O emissions were fitted to a linear variogram in 1999 and responded to a spherical variogram model in 2000. Positive first degree surface trends were also found for N_2O emissions data in both years. However, the removal of these trends did not change variogram models, but significantly improved them by increasing the R^2 and Q values. N_2O emissions systematically varied with small zones of uptake (negative flux) across the field, suggesting the presence of hot spots.

Key words: Nitrous oxide emissions, spatial variability, lowland soil

INTRODUCTION

The tropospheric concentration of nitrous oxide (N_2O), a potent greenhouse gas also involved in the catalytic degradation of stratospheric ozone, has increased since the beginning of the Industrial Era, with a rate of 0.8 ppb per year during the 1990s (Dambreville *et al*., 2008). Nitrous oxide (N_2O) is a trace gas that has received considerable attention because of its importance in atmospheric chemistry and its influence in controlling the global heat budget (IPCC, 2007). Anthropogenic activities such as agriculture have been named among the major causes for the increase of this pollutant (Lee *et al*., 2006; Mosier *et al*., 1998). In fact, Both N_2O and CH_4 are the most prominent GHGs in agriculture and their emissions amount to approximately 39% of global N_2O emissions and 40% of global CH_4 emissions (Oenema *et al*., 2001). That is why a series of best management practice options have been suggested to mitigate the emission of CO_2, N_2O and CH_4 from agriculture and to have agriculture serve as a sink for atmospheric GHG (Desjardins *et al*., 2001; Follett, 2000; Oenema *et al*., 2001; Paustian *et al*., 2000). Unfortunately, the high degree of spatial variability of N_2O (as well as other greenhouse gases)

Corresponding Author: Nsalambi V. Nkongolo, Environmental Science Program,
Department of Cooperative Research, College of Natural Sciences,
Mathematics and Technology and Cooperation Research and Extension,
Lincoln University of Missouri, Jefferson City, MO 65102-0029, USA

emissions and soil-controlling soil properties still presents a major challenge to accurately quantifying fluxes (Chatskikh and Olesen, 2007; Ruser *et al*., 2006; Weitz *et al*., 2001; Flessa *et al*., 2002; Rover *et al*., 1999). In fact, N_2O fluxes across geographic regions vary in response to major and repetitive differences in the soil environment (Lamers *et al*., 2007; Chao *et al*., 2000; Mummey *et al*., 1998). For example, at a micro-to-plot scale, N_2O fluxes are controlled by the availability of soil water, labile C and inorganic N. These factors are, in turn influenced by soil type and plant community type at the landscape scale. The distributions of soil types and plant communities are interrelated and their variation in a region is again largely controlled by geomorphology, land use and climate. Therefore, quantification of N_2O flux rates from different sites, ecosystems, crops, climate and agricultural practices is necessary to improve the accuracy of N_2O emissions inventories (Lamers *et al*., 2007; Boeckx and Van Cleemput, 2006; Chao *et al*., 2000; Mummey *et al*., 1998). The objective of this study was to assess the spatial and temporal variability of N_2O emissions in a field cropped to onion.

MATERIALS AND METHODS

Study Area

Air and soil samples for determination of N_2O emissions and soil properties, respectively, were collected in Mikassa, Hokkaido province (Fig. 1), but all analyses were done in the Laboratory of Soil Science at Hokkaido University in Sapporo. Sapporo is Japan's third largest city in area and is located on the western plains of Hokkaido, the northernmost island of Japan. Its geographical locations are 43°11′N, 141°30′E. Sapporo enjoys a mild climate with a year-round average temperature of 9.1°C. The average temperature in January was -3.7°C and in July, 20.3°C in 2000. More than sixty percent of surface area of Sapporo (primarily in the southwest) is mountainous, creating a concentration of urban activity focused around the Toyohira River, which runs through the city.

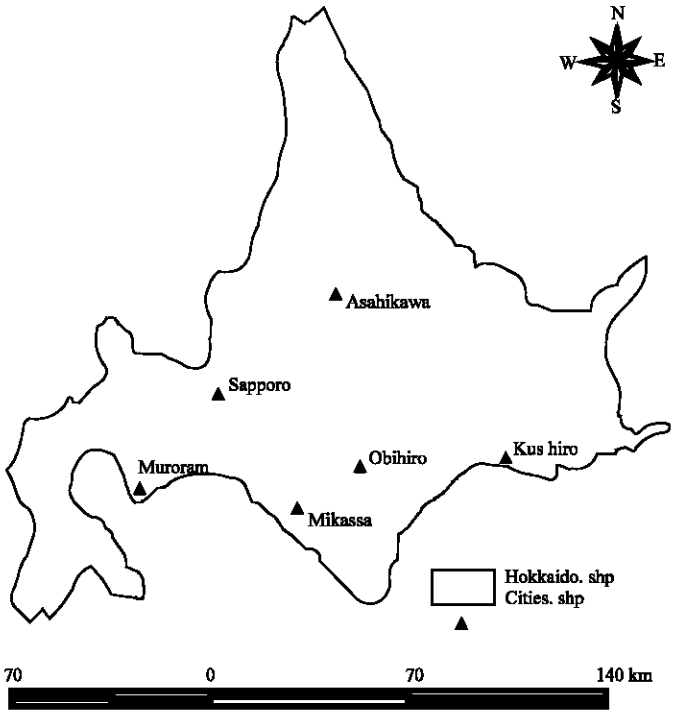

Fig. 1: Hokkaido, Japan

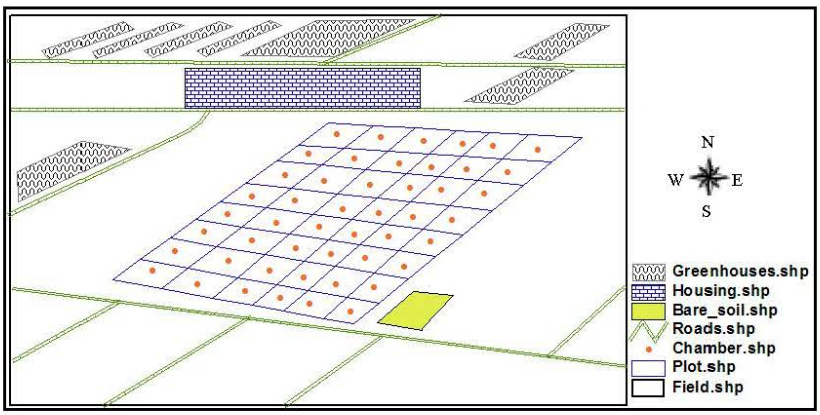

Fig. 2: Experimental field in Mikassa, Hokkaido (Japan)

Experimental Field

The study was conducted in a 140 by 140 m upland field yearly cropped to onion (*Allium cepa* L.) in Mikassa, Hokkaido, Japan (43°14'N, 141°50'E). The experimental field is showed in Fig. 2. The annual average temperature in Mikassa is 7.2°C and the average annual rainfall is 1204 mm. The soil of the experimental site is classified as fine, mesic, mollic Fluvaquent. The physical and chemical properties of different horizons were reported elsewhere. Soil texture consists of a silty or heavy clay from the Ap layer (0-28 cm) down to the C horizon (48-100+ cm). The groundwater table lays at 70-80 cm depth throughout the growing season (Hu *et al*., 2004). Surface drains were installed at 80-100 cm depth at 12 m intervals and were connected to the same effluent exit, draining about 0.95 ha (125 by 76 m) for monitoring nitrate leaching. Fertilizer nitrogen (322 kg N ha^{-1}) was applied at the end of April, shortly before transplanting. Onion was harvested during the second and third week of September. In June 1999, the field was sampled for N_2O emissions and soil physical and chemical properties, using a 100 by 100 m grid at 10 m spacing for a total of 100 sampling locations. A year later in September 2000, the same field was again sampled for N_2O and soil chemical and physical properties, using a 60 by 60 m grid at 10 m spacing for a total of 36 locations. Data collected in both 1999 and 2000 are analyzed in this study.

Measurements of N_2O Emissions

N_2O emissions from the soil surface, which may have been produced in the root zone or in deeper horizons, were measured using a closed-chamber technique. The chambers were circular with steel frames. The top of each chamber had a gas sampling tube and a bag to control air pressure inside the chamber. The height and diameter of the chamber were 0.35 and 0.30 m, respectively. At each sampling time, 6 chambers were installed into the soil and kept for 20 min and then samples of the enclosed atmosphere were withdrawn by a 50 mL syringue and transferred into a 1 L Tedlar ® Bag. The air temperature inside the chamber was recorded using a digital thermometer. Ambient air between 0 and 2 m from the soil surface was collected and its mean concentration was used as a background concentration for calculation of gas fluxes. A gas chromatography with electron capture detector (Shimadzu, model 14°C) was used for N_2O analysis. The gas flux was calculated according to Ginting *et al*. (2003):

$$F = \rho * \frac{V}{A} * \frac{\Delta C}{\Delta t} * \left(\frac{273}{T}\right) * \alpha \quad (1)$$

where, F is the gas production rate (μg N_2O-N m^{-2} h^{-1}); ρ is the gas density (mg m^{-3}) under standard conditions; V (m^3) and A (m^2) are the volume and bottom area of the chamber; $\Delta C/\Delta t$ is the ratio of change in the gas concentration inside the chamber (10^{-6} m^3 m^{-3} h^{-1}); T is the absolute temperature and α is the transfer coefficient (28/44 for N_2O). A positive value indicates gas emission from the soil, while a negative value indicates gas uptake. The detectable limit was 0.1 ug N m^{-2} h^{-1}. Soil temperature was measured at 5 and 10 cm from the top soil layer, using a digital thermometer.

Geostatistical and Statistical Analyses

Data Detrending

Basic assumptions of regionalized variable studies are often overlooked or rarely verified by many researchers. This may introduce artifacts and confound the interpretation of results. Therefore it is important to conduct a thorough data analysis before and during geostatistical analysis to filter out site-specific potential problems (producing trends) related to the unknown regionalized variable (i.e., soil gas flux under investigation). We used the median polishing technique to clean (polish) our field data to satisfy the basic assumptions for the estimation of a semivariogram, that is, for second-order stationarity or the weaker intrinsic hypothesis. Second-order stationarity implies that the mathematical expectation E[Z(x)] = μ exists and does not depend upon the position x and that for each pair of regionalized variables [Z(x), Z(x+h)], the covariance exists and depends only upon the separation vector h. On the other hand, the weaker intrinsic hypothesis implies that the mathematical expectation E[Z(x)] = μ exists and for all vectors *h* the increment [Z(x+h)-Z(x)] has a finite variance that does not depend on x (Journel and Huijbregts, 1978). The methodology consisted in first determining whether there was a statistically significant linear or polynomial trend surface to the data. The procedure consisted in solving either a linear or a second order trend surface equation. Using Mathcad 4.0 software, the linear trend surface equation was developed and solved as follows:

$$Y = b_0 + b_1X_1 + b_2X_2 \tag{2}$$

where, Y, the field measured gas flux or soil property is regarded as a linear function of some constant value (b_0) related to the means of observation, plus an east-west (b_1) coordinate component and a north-south (b_2) component. Since this equation contains three unknowns, three normal equations were needed to find its solution:

$$\begin{aligned}
\sum Y &= b_0 n + b_1 \sum X_1 + b_2 \sum X_2 \\
\sum X_1 Y &= b_0 \sum X_1 + b_1 \sum X_1^2 + b_2 \sum X_1 X_2 \\
\sum X_2 Y &= b_0 \sum X_2 + b_1 \sum X_1 X_2 + b_2 \sum X_2^2
\end{aligned} \tag{3}$$

Solving this series of simultaneous equations with Mathcad software gave the coefficients of the best linear trend surface.

$$\begin{bmatrix} n & \sum X_1 & \sum X_2 \\ \sum X_1 & \sum X_1^2 & \sum X_1X_2 \\ \sum X_2 & \sum X_1X_2 & \sum X_2^2 \end{bmatrix} \cdot \begin{bmatrix} b_0 \\ b_1 \\ b_2 \end{bmatrix} = \begin{bmatrix} \sum Y \\ \sum X_1Y \\ \sum X_2Y \end{bmatrix} \tag{4}$$

For the second-degree trend surface, the following equation was used:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_1^2 + b_4X_2^2 + b_5X_1X_2 \tag{5}$$

The equation contains terms that are the squares of the two geographic coordinates (X_1 for X and X_2 for Y) and a cross product term X_1X_2 Because its contains six unknowns, six normal equations were developed. The solution of these equations gave the coefficients of the best fit:

$$\begin{bmatrix} n & \sum X_1 & \sum X_2 & \sum X_1^2 & \sum X_2^2 & \sum X_1X_2 \\ \sum X_1 & \sum X_1^2 & \sum X_1X_2 & \sum X_1^3 & \sum X_2^2X_1 & \sum X_1^2X_2 \\ \sum X_2 & \sum X_2X_1 & \sum X_2^2 & \sum X_1^2X_2 & \sum X_2^3 & \sum X_2^2X_1 \\ \sum X_1^2 & \sum X_1^3 & \sum X_1^2X_2 & \sum X_1^2 & \sum X_1^2X_2^2 & \sum X_1^3X_2 \\ \sum X_2^2 & \sum X_2^2X_1 & \sum X_2^3 & \sum X_2^2X_1^2 & \sum X_2^2 & \sum X_2^3X_1 \\ \sum X_1X_2 & \sum X_1^2X_2 & \sum X_1X_2^2 & \sum X_1^3X_2 & \sum X_2^3X_1 & \sum X_1^2X_2^2 \end{bmatrix} \begin{bmatrix} b_0 \\ b_1 \\ b_2 \\ b_3 \\ b_4 \\ b_5 \end{bmatrix} = \begin{bmatrix} \sum Y \\ \sum X_1Y \\ \sum X_2Y \\ \sum X_1^2Y \\ \sum X_2^2Y \\ \sum X_1X_2Y \end{bmatrix} \quad (6)$$

When significant trend was found, the field data matrix was detrended as follows:

$$D_{ij} - T_{ij} = R_{ij} \quad (7)$$

where, D_{ij} is the original data matrix for points ij, T_{ij} is the trend surface at points ij and R_{ij} is the matrix of residuals, i is the column and j the corresponding row. The subsequent geostatistical analysis was performed on the residual matrix. The map developed by kriging using the results of the geostatistical analysis of the residual matrix was then added to the trend surface to determine the final maps of gas fluxes and soil properties. The examination of the trend surface determined whether universal (with trend surface) or ordinary (no trend surface) kriging was to be performed.

Variograms Fitting

Isotropic (direction independent) semivariance of data was calculated using GS^+ geostatistical software (Gamma Design Software, 2007). Semivariance is defined in the following equation:

$$\gamma(h) = \frac{1}{[2m(h)]} \sum_{i=1}^{m(h)} \left[Z_{(xi)} - Z_{(xi+h)} \right]^2 \quad (8)$$

where, γ is the semivariance for m data pairs separated by a distance of h, known as a lag and Z is the value at positions xi and xi+h.

Statistical Analysis

Statistix 8.0 was used for computing summaries of simple of statistics and well as histograms.

RESULTS AND DISCUSSION

Summary of Simple Statistics for Nitrous Oxide Emissions

Histograms for N_2O emissions and their transformation into logarithmic scale are showed in Fig. 3 and 4 (a and b) for 1999 and 2000, respectively. Descriptive statistics for nitrous oxide emissions in Mikassa in June 1999 and September 2000 are given in Table 1. Overall, N_2O emissions in June 1999 were highest as compared to values obtained in September 2000. The mean value of N_2O emissions in June 1999 was almost two times higher as compared to that measured in September 2000. However, both means become statistically different when data are transformed into logarithmic scale. The difference in N_2O emissions between these two years of studies can be explained by several factors such as the month of sampling within each year: June 1999 versus September 2000; the number of samples collected each year: 100 samples in 1999 versus 36 samples in 2000 and by soil properties

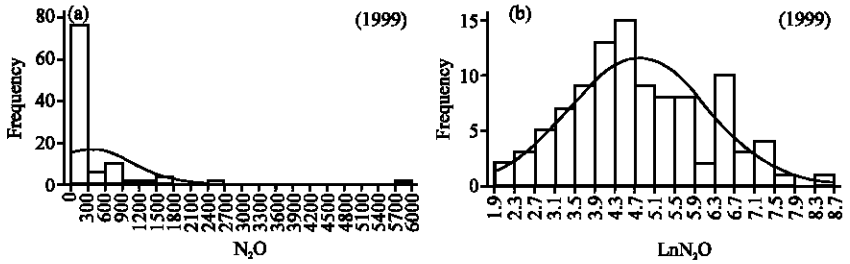

Fig. 3: Histograms of N_2O emissions in 1999: (a) original data and (b) log-transformed (Ln) data

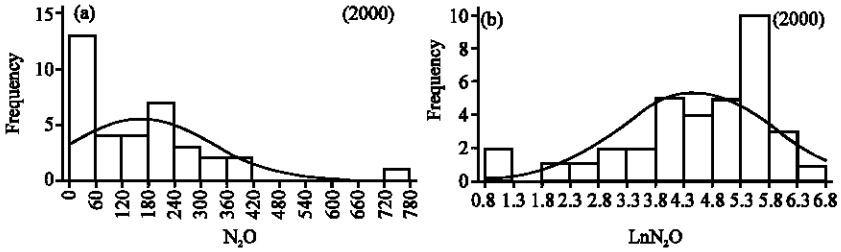

Fig. 4: Histograms of N_2O emissions in 2000: (a) original data and (b) log-transformed (Ln) data

Table 1: Summary of simple statistics for nitrous oxide (N_2O) emissions in Mikassa (Japan)

	1999		2000	
Simple statistics	N_2O ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	Ln (N_2O) ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	N_2O ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	Ln (N_2O) ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)
Mean	313.41	4.72	165.26	4.52
SD	704.72	1.38	155.24	1.34
CV	224.86	29.39	93.94	29.55
Median	94.35	4.55	148.47	4.99
Skew	5.85	0.29	1.78	0.99

such as soil temperature. With particular attention to soil temperature, during field sampling in June 1999, the average soil temperatures at 5 and 10 cm depth from the soil surface (our sampling depth) were 30.99 and 28.04°C, respectively (data not showed). However, in September 2000, the soil temperature at these two depths dropped to 17.53 and 15.91, respectively. These results agree with those of Hu *et al.* (2001) who also reported an increase in N_2O emissions as a result of increasing soil temperature. In fact, soil temperature has been reported as one of the factors influencing the seasonal variability of nitrous oxide emissions. Weiss and Price (1980) suggested that N_2O solubility generally increases as the solution temperature decreases, this implies that during fall and winter, the N_2O emitted from soil could be lower than the actual N_2O produced in the soil because part of the N_2O stayed in the soil solution. As for the means, coefficients of variation (CV) were also highest in 1999 as compared to 2000, confirming that N_2O emissions exhibited considerable spatial and temporal variability. In fact, temporal patterns in gaseous N losses have been described by various workers. These patterns have differed according to ecosystem and geographic location (Lemke *et al.*, 1998). In a denitrification study at a forest site in Michigan, Groffman and Tiedje (1989) found that gaseous N fluxes were highest in early spring and late fall with negligible values recorded during summer. Van Kessel *et al.* (1993) found highest emissions of N_2O during early spring and summer with fluxes declining to negligible levels in the late growing season and fall. Nyborg *et al.* (1997) reported high

Table 2: First degree surface trend analysis for N_2O emissions in Mikassa (Japan)

	1999		2000	
Analysis variable	N_2O ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	Ln (N_2O) ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	N_2O ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	Ln (N_2O) ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)
r	0.43	-	0.44	-
F value*	10.99	-	3.97	-

*Critical F value = 2.56

Table 3: Isotropic semivariogram parameters for N_2O and their de-trended residuals

	1999		2000	
Par.	N_2O ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	Residuals ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	N_2O ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)	Residuals (N_2O) ($\mu g\ N_2O\text{-}Nm^{-2}\ h^{-1}$)
Model	LIN	LIN	SPH	SPH
Nugget	516.00	100.00	1.00	1.00
Sill	1442.95	115390.51	1665.00	2873.00
Range	61.32	61.32	16.48	24.97
Q	0.64	0.99	0.99	1.00
R^2	0.68	0.94	0.45	0.90

LIN = Linear, SPH = Spherical

fluxes of N_2O during spring thaw, but negligible flux during the following growing season at an agricultural site in Alberta.

Surface Trends Analysis for Nitrous Oxide Emissions

Results of first degree surface trend analysis is shown in Table 2. There was a significant first degree trend analysis for N_2O emissions measured in both years. However, the trend was more prevalent in 1999 in comparison to 2000 as showed by the higher probability value of 10.99 observed in 2000 data analysis. Log-transformed data did not, however, show any significant trend.

Variogram Models Fitting for Nitrous Oxide Emissions

Isotropic semivariogram parameters for field measured N_2O emissions and their detrended residuals are shown in Table 3 for both 1999 and 2000, respectively. Figure 5 and 6 (a and b) show semivariogram models fitted to data in both years. The criterion for model selection was maximum R^2, except in cases where another model was obviously more appropriate based on visual examination of the semivariogram. Neither an active lag distance nor a lag interval was set. Default values given by the program were used. A linear variogram was fitted to data in 1999 and a spherical model was more appropriate for N_2O emissions data in 2000. This applies to both field measured data and detrended residuals. With isotropic models of field data, the ranges or limits of spatial dependency were 61.32 and 16.48 m for 1999 and 2000, respectively. Detrended residuals for 1999 had the same limit of spatial dependency as field measured data. However, this situation changed in 2000 when the limit of spatial dependency for detrended residuals exceeded that of field measured data. In both cases, the limit of spatial dependency was lower than the sampling distance, which is usually accepted. The sill values for field measured N_2O were lowest as compared to those obtained with detrended data. The Q value for field measured N_2O fluxes in 1999 was 0.68 and it approached unity (0.99) in 2000, suggesting a highly developed spatial structure for N_2O fluxes in 2000 and a moderate development of spatial structure in 1999. Detrended residual Q values were also very high, showing highly developed spatial structure for both years. In opposite to the Q values trend the R^2 for field measured N_2O fluxes were higher in 1999 and lower in 2000. However, when data were detrended the R^2 became high for both year, suggesting that removing the trend was useful in the analysis of this data. Figure 2 shows the semivariograms of N_2O fluxes in 1999 and 2000 for both original field data and detrended residuals. A

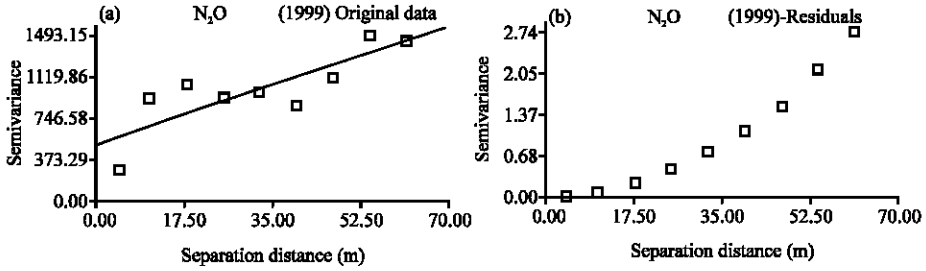

Fig. 5: Variogram of N_2O emissions in 1999: (a) Original data and (b) De-trended Residuals

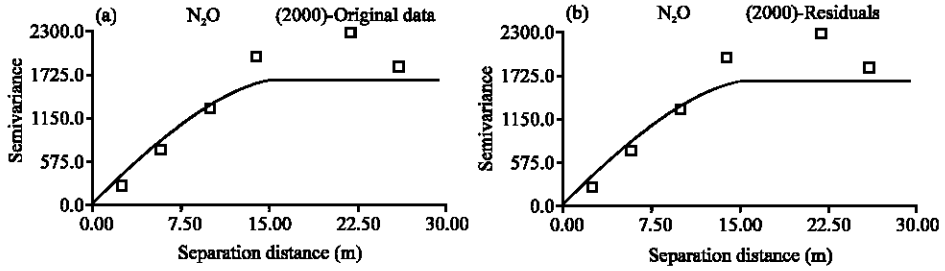

Fig. 6: Variogram of N_2O emissions in 2000: (a) Original data and (b) De-trended residuals

linear variogram model was fitted to N_2O fluxes in 2000 (Fig. 5a, b) with an active lag distance of 100 m. As the figure clearly shows, multiple spatial scales seemed to be present in this isotropic variogram with evidence of a trend. After detrending the data, the isotropic variogram exhibited a linear trend with the same range, but with higher Q and R^2 and an improved fit. For the 2000 (Fig. 6a, b) data, a spherical trend was fitted to the data with an active lag of 60 m. After detrending the data, a better fit was obtained with not only increased Q and R^2, but the range of spatial dependence also increased.

Mapping Nitrous Oxide Emissions Across the Field

Maps of N_2O emissions distribution in an onion field in Mikassa in 1999 and 2000 with first degree trend surface and contoured residuals are shown in Fig. 7 and 8a-c. Estimation of N_2O emissions at unsampled locations was made using the geostatistical technique of kriging. A default grid spacing was used for interpolation purposes. After estimation of N_2O emissions, isarithmic maps were produced with different contour levels. Maps showed that there is a systematic variability with spatial patterns of N_2O emissions in the onion field. These patterns also differ for each year. In fact, Fig. 7a shows that in 1999, field measured N_2O emissions were nearly homogenous across the onion field with spots of higher values in the western part of the onion field. A small zone of N_2O uptake (negative flux) was also observed in the southwestern part of the onion field. However, maps produced after detrending the data (Fig. 7c) showed a different picture: several zones of N_2O emissions spots with spots of higher values in northwest but also southern parts of the onion field. However, the small zone of uptake has moved in northwestern part of the onion field. Maps of N_2O emissions in 2000 showed more variability. For field collected data, a large zone of higher N_2O emissions was observed in northwestern, moved to the middle and extended to east and southern parts of the onion field. Two spots of N_2O uptake were also found in the western part of the onion field. Detrending data did not change much of the variability of N_2O emissions across the onion field in 2000. In fact, a large zone

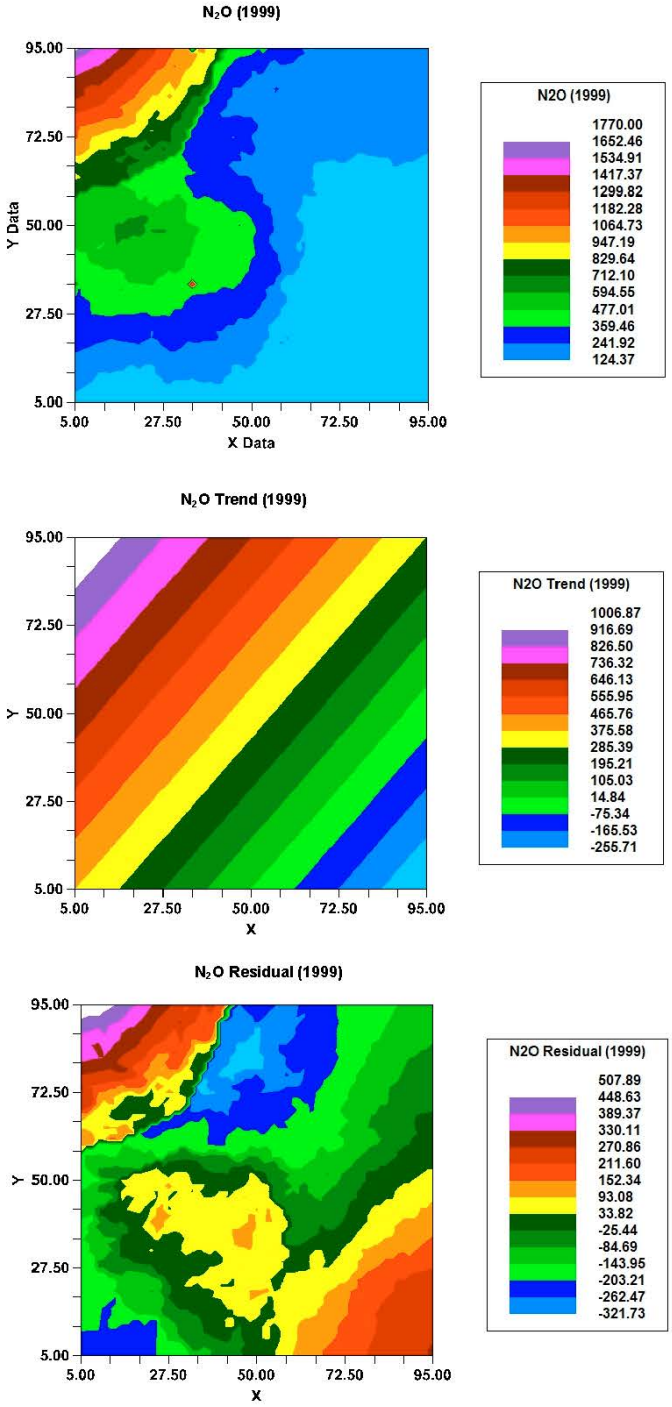

Fig. 7: N_2O emissions in Mikassa in 1999: (a) Contour maps, (b) First degree trend surface and (c) Contoured residuals from first degree trend

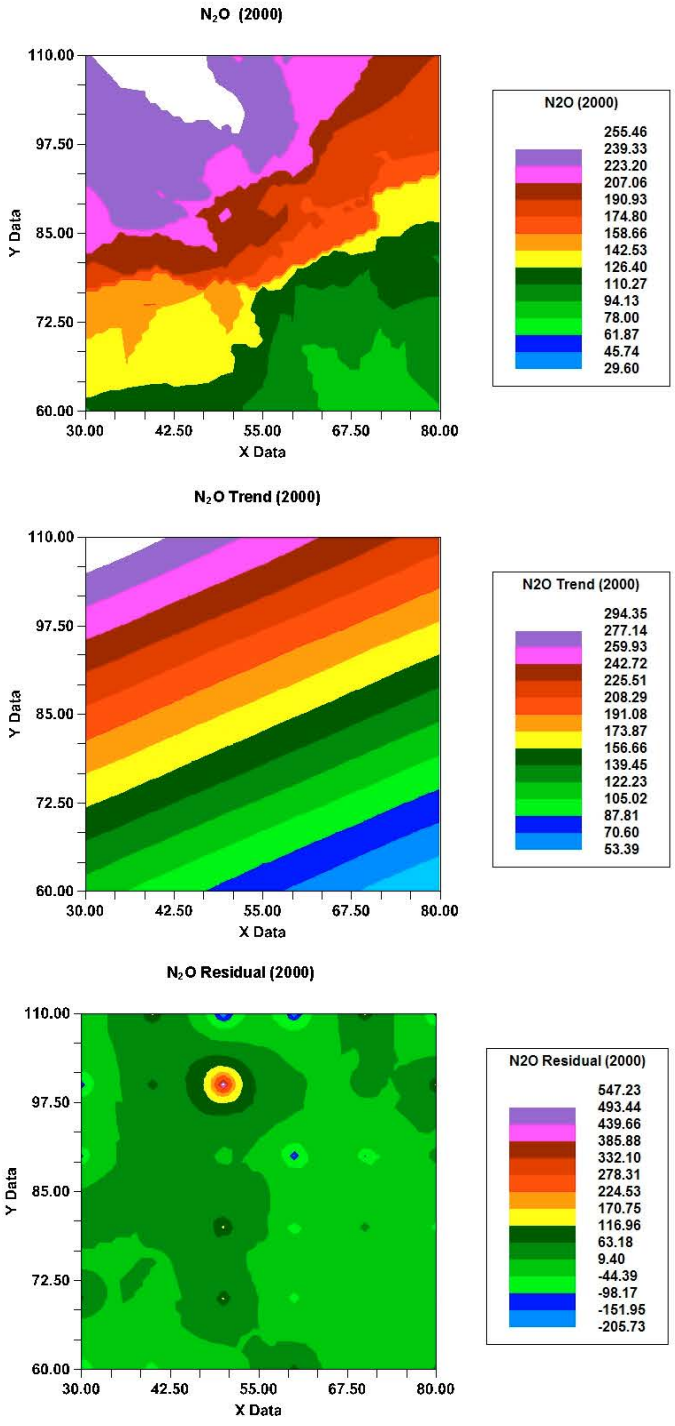

Fig. 8: N_2O emissions in Mikassa in 2000: (a) Contour maps, (b) First degree trend surface and (c) Contoured residuals from first degree trend

of higher N_2O emissions is still observed in northern, middle and southern parts of the onion field. However, only one spot of uptake is found and concentrated in northwestern part of the onion field (Fig. 8a).

CONCLUSION

This study was conducted to assess the spatial variability of N_2O fluxes in a field cropped to onion. Results obtained showed that N_2O emissions were highest in 1999 as compared to 2000. They were fitted to a linear variogram in 1999 while they responded to a spherical variogram model in 2000. Positive first degree surface trends were also found in N_2O emissions data in both years and the removal of these trends did not change variogram models, but significantly improved them by increasing the R^2 and Q values. N_2O emissions systematically varied with small zones of uptake (negative flux) across the field. This study is another confirmation of the tremendous spatial and temporal variability of N_2O emissions. Variability of N_2O emissions in space may be influenced by site-specific potential problems (producing trends) related to the unknown regionalized variable (i.e., soil property under investigation). Therefore, removal of a potential trend was used in this study to improve variogram fitting. However, the results showed that in many cases, the variogram fitted to data after trend removal had poor spatial structure and low R^2. In some cases however, both the spatial structure and R^2 improved. N_2O emissions systematically varied with small zones of uptake (negative flux) across the field, suggesting the presence of hot spots.

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