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Nitrous Oxide (N_2O) Emissions from a Japanese Lowland Soil Cropped to Onion: II. Relationship with Soil Chemical Properties

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Abstract: The spatial variability of soil chemical properties and their relationships with N_2O emissions were studied in a lowland soil cropped to onion in Mikassa, Hokkaido (Japan). Soil air for the determinations of N_2O emissions and soil samples for chemical properties analyses were collected in 100 by 100 m and 60 by 60 m grids in 1999 and 2000, respectively with samples taken at 10 m spacing. Soil chemical properties studied were pH, electrical conductivity (EC), nitrite (NO_2^-), nitrate (NO_3^-), ammonia (NH_4^+), sulfate (SO_4^{2-}), phosphate (PO_4^{3-}), chloride (Cl^-), total carbon (TC), C/N ratio and total nitrogen (TN). Results showed that in both years, soil chemical properties fitted to either linear, exponential or spherical variogram models. In 1999, soil chemical properties showed positive first degree surface trends, but removing them resulted in decreasing the range of spatial variability (Ao) and R^2 values. However, there was no positive trend in 2000. N_2O emissions were directly related with NO_2^-, PO_4^{3-} and soil temperature. However, correlations between other soil chemical properties and N_2O emissions were possible only after transforming the data into logarithmic scale. The highest correlations were obtained between N_2O emission and PO_4^{3-} ($p = 0.0001$, $r = 0.61$), followed by the sum of N-radicals $NO_3^- + NH_4^+$ ($p = 0.003$, $r = 0.48$). This study confirms that, besides those traditionally studied, other soil chemical properties might as well correlate with N_2O emissions. In depth studies of the relationship between N_2O emissions and these soil chemical properties are needed.

Key words: Spatial variability, soil chemical properties, N_2O emissions

INTRODUCTION

Soils have been identified as the dominant source of N_2O, contributing about 57% (9Tg per year) of the total annual global emissions, of which about 27% (2.4Tg per year) originates from agricultural soils (Jones *et al*., 2007). The mechanisms responsible for N_2O emissions from soils are the microbial processes of (a) nitrification, the oxidation of ammonium to nitrate, (b) denitrification, the anaerobic reduction of nitrate to gaseous forms of N (Mosier *et al*., 1998). Both processes are controlled by the availability of mineral N, soil temperature, pH, moisture, texture, organic carbon, the availability of soluble organic matter and other factors (Skiba and Smith, 2000). Several of these factors are in turn

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controlled by soil and crop management practices (Grant *et al*., 2004; Dobbie and Smith, 2003a, b; Dobbie and Smith, 2001; Goossens *et al*., 2001; Anger *et al*., 2003; Sitaula *et al*., 2000a, b; Wild *et al*., 1998; Flessa *et al*., 2002a, b; Wang *et al*., 2005; Yamulki and Jarvis, 2002; Teepe *et al*., 2004). Large spatial and temporal variability is therefore typical for these emissions and coefficient of variation higher than 100% are not uncommon (Jacinthe and Lal, 2006; Yanai *et al*., 2003; Röver *et al*., 1999). This high degree of spatial and temporal variability of nitrous oxide emissions (and other greenhouse gases) and soil controlling factors present a major challenge to accurately quantifying emissions from agricultural fields (Houghton *et al*., 2001). Matson *et al*. (1989) suggested that our ability to quantify greenhouse gas emissions and especially to extrapolate to larger scales without making vast numbers of direct flux measurements should improve with improved knowledge of the soil enviroumental factors governing emissions. By assessing the spatial and temporal variability of emissions in relation to potential soil-controlling factors, identification of the most influential factors should be possible and such information would be particularly useful in predictive emission models (Grant *et al*., 1993). There are several potential soil controlling factors, but only a few of them, such as soil water (water-filled porosity) and indices of nitrogen availability (NO_3^- and NH_4^+) have so far received much attention (Saggar *et al*., 2007; Jones *et al*., 2007). The objective of this study was to assess the spatial variability of soil chemical properties. A second objective was to study the relationship between soil chemical properties and 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 in part I), 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 60% 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.

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 Fig. 2, part I of this study. 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. Surface drains are installed at 80-100 cm depth at 12 m intervals and are 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}) is yearly applied once at the end of April, shortly before transplanting. Onion is harvested during the second or 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 1999 and 2000 are analyzed in this study.

Measurements of Soil Chemical Properties

Two cores corresponding each to 100 cm^3 of soil were taken at 5 cm depth at each sampling locations after N_2O emissions measurements, for analysis of soil pH (H_2O and KCl), Electrical Conductivity (EC), chloride (Cl^-), nitrite (NO_2^-), nitrate (NO_3^-), ammonium (NH_4^+), sulfate and phosphate. For analyzing soil Cl^-, NO_2^- and NO_3^-, 10 g of field moist soil sample were extracted by 50 mL of deionized water (1:5 = soil:water) and concentrations of the above anions were determined by ion exchange chromatography. The above extract was also used to measure pH (H_2O) and EC. For NH_4^+ determination, 7 g of field moist sample was extracted using 70 mL of KCl 2 M. pH (KCl) was measured using this extract and soil NH_4^+ was determined by colorimetry with indophenol-blue.

Spatial and Statistical Analysis

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:

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

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.

RESULTS AND DISCUSSION

Analysis of the Spatial Variability of Soil Chemical Properties

Summary of Simple Statistics for Soil Chemical Properties

Summary of simple statistics for soil chemical properties from an agricultural field cropped to onion in Mikassa, Hokkaido (Japan) is shown in Table 1 for June 1999 and September 2000, respectively. Many chemical properties measured in June 1999 were not measured in September 2000 and vice-versa. Ouly soil pH and Electrical Conductivity (EC) were measured in both years, this in addition to other soil chemical properties. Overall, coefficients of variation (CV) ranged from 38 to 261% in 1999 while they ranged from about 5 to 100% in 2000. It seems that the high variability in soil chemical properties in 1999 resulted in high variability of N_2O emissions in 1999. In fact,

Table 1: Summary of simple statistics for soil chemical properties measured in anion field in Mikassa (Japan)

	1999					2000				
Properties	Mean	SD	CV (%)	Med.	Sk.	Mean	SD	CV (%)	Med.	Skew
pH (H_2O)	7.59	19.84	261.28	5.61	9.84	6.00	0.29	4.83	6.07	-0.34
pH (KCl)	-	-	-	-	-	4.61	0.39	8.50	4.53	1.59
EC (mS)	2.35	1.45	61.52	1.85	1.59	5.80	1.60	27.67	5.44	2.12
Cl^- (mg Cl kg^{-1})	-	-	-	-	-	7.63	2.33	30.61	7.55	1.72
PO_4^{-3} (mg P kg^{-1})	-	-	-	-	-	16.63	6.89	41.40	15.58	0.65
SO_4^{-2} (mg S kg^{-1})	-	-	-	-	-	21.12	15.13	71.64	17.08	1.85
NO_2^- (mg N kg^{-1})	-	-	-	-	-	0.08	0.08	99.56	0.07	2.24
NO_3^- (mg N kg^{-1})	-	-	-	-	-	10.15	5.31	52.32	8.98	0.87
NH_4^+ (mg N kg^{-1})	-	-	-	-	-	1.10	0.34	30.84	1.10	-0.21
C/N	13.72	1.55	11.28	13.80	-6.53	-	-	-	-	-
TN	0.23	0.34	142.90	0.21	9.74	-	-	-	-	-
TC	2.91	1.11	37.99	2.82	8.62	-	-	-	-	-

pH = Potential hydrogen, EC = Electrical conductivity, Cl^- = Chloride, C/N = Carbon-nitrogen ratio, TN = Total nitrogen, TC = Total carbon, NO_2^- = Nitrite, NO_3^- = Nitrate, NH_4^+ = Ammonia, PO_4^{-3} = Phosphate, SO_4^{-4} = Sulfate

Table 1 shows that CV for N_2O emissions in 1999 was 224.86%, a value twice as much as that observed in 2000. This high variability observed in 1999 has also resulted high skewness values, which ranged from 1.59 to 9.84 in 1999 while they ranged only between 0.21 to 2.24 in 2000. In addition, mean values for soil chemical properties measured in 2000 were closer to their corresponding medians with low SD (except for soil sulfate), suggesting that data in 2000 better approached normal distribution despite the fact that the sample size was nearly 3 times less than in 1999. The high variability in soil chemical properties can be explained by reasons stated earlier regarding the variability of N_2O emissions. As said earlier, in June 1999, the average soil temperatures at 5 and 10 cm depth from the soil surface were 30.99 and 28.04°C, respectively. However, in September 2000, the soil temperature at these two depths dropped to 17.53 and 15.91°C, respectively. Since the speed of a chemical reaction increases with increasing soil temperature, it can be assumed that more reactions occurred in the soil in 1999, leading to higher CV values.

Surface Trends Analysis for Soil Chemical Properties

Table 2 shows the linear trend analysis for soil chemical properties in 1999 and 2000 in Mikassa, Hokkaido (Japan). All soil chemical properties measured in 1999 showed a significant linear trend while there was no significant trend in 2000. Total carbon (TC) and total nitrogen (TN) had the most significant linear trend with the highest coefficients of correlation (r) and F-values.

Variogram Models Fitting for Soil Chemical Properties

Isotropic variogram parameters for soil chemical properties are showed in each variogram's figure but are also shown in Table 3 and 4 for 1999 and 2000, respectively. Variogram parameters for

Table 2: Linear trend analysis for soil chemical properties measured in an onion field in Mikassa (Japan)

	1999		2000	
Properties	r	F	r	F
pH (H_2O)	0.54	20.10	0.29	1.55
pH (KCl)	-	-	0.16	0.41
EC (mS)	0.24	2.90	0.33	1.95
Cl^- (mg Cl kg^{-1} soil)	-	-	0.12	0.25
PO_4^{-3} (mg P kg^{-1} soil)	-	-	0.16	0.41
SO_4^{-2} (mg S kg^{-1} soil)	-	-	0.30	1.62
NO_2^- (mg N kg^{-1} soil)	-	-	0.12	0.26
NO_3^- (mg N kg^{-1} soil)	-	-	0.06	0.00
NH_4^+ (mg N kg^{-1} soil)	-	-	0.21	0.78
C/N	0.57	23.77	-	-
TN	0.87	148.38	-	-
TC	0.83	105.88	-	-

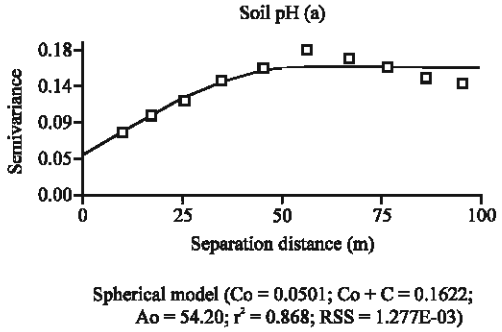
Critical F-value = 2.56

Table 3: Isotropic variogram parameters and their detrended residuals for soil chemical properties measured in an onion field in Mikassa (Japan) in June 1999

	Field measured properties					
Properties	Model	Nugget	Sill	Range	C/(Co+C)	R^2
pH (H_2O)	SPH	0.05	0.16	54.20	0.69	0.87
EC (mS)	SPH	10.00	4221.00	46.10	0.99	0.99
C/N	SPH	0.21	1.62	310.90	0.87	0.92
TN	GAU	0.00	0.02	260.80	0.99	0.97
TC	GAU	0.02	1.90	261.30	0.99	0.97
Detrended residuals						
pH (H_2O)	SPH	0.00	0.02	32.90	0.00	0.00
EC (mS)	LIN	2293.00	2293.00	95.79	0.99	0.50
C/N	SPH	0.00	0.07	50.80	0.99	0.95
TN	LIN	0.00	0.00	95.28	0.00	0.11
TC	EXP	0.03	0.06	310.90	0.50	0.12

Table 4: Isotropic variogram parameters and their detrended residuals for soil chemical properties measured in an onion field in Mikassa (Japan) in September 2000

	Field measured properties					
Properties	Model	Nugget	Sill	Range	$C/(C_o+C)$	R^2
pH (H_2O)	SPH	0.00	0.12	36.45	1.00	0.98
pH (KCl)	SPH	0.00	0.12	29.83	1.00	0.98
EC (mS)	SPH	0.00	0.08	17.45	1.00	0.72
Cl^- (mg Cl kg^{-1} soil	EXP	0.03	0.24	22.17	0.87	0.75
PO_4^{-3} (mg P kg^{-1} soil)	LIN	0.18	6.90	34.06	0.97	0.98
SO_4^{-2} (mg S kg^{-1} soil)	SPH	0.10	38.73	26.48	0.99	0.95
NO_2^- (mg N kg^{-1} soil)	LIN	0.00	0.01	34.06	1.00	0.92
NO_3^- (mg N kg^{-1} soil)	SPH	0.01	3.41	9.66	0.99	0.36
NH_4^+ (mg N kg^{-1} soil)	LIN	0.00	0.03	34.06	0.97	0.89



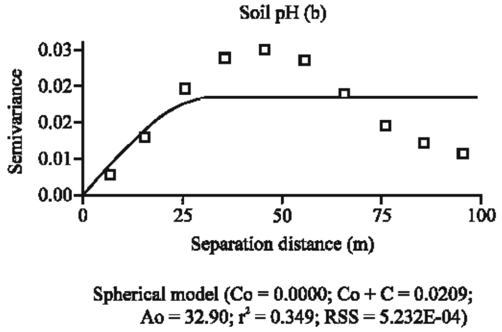
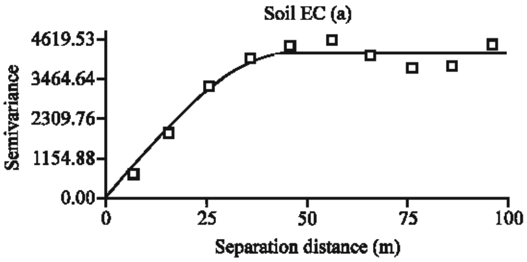
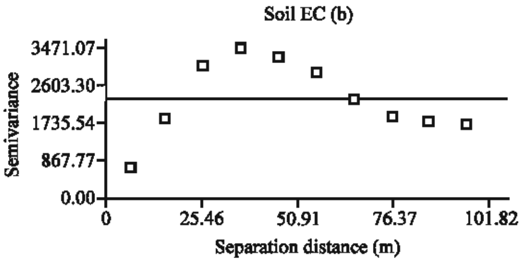


Fig. 1a: Variograms of soil pH: (i) original data and (ii) de-trended residuals

detrended residuals are showed in Table 3 only because there was no significant trend in soil chemical properties in 2000. The corresponding variograms are shown in Fig. 1a-c and 2a, b. In 1999, soil pH, EC and C/N responded to spherical variogram models while TN and TC were fitted to gaussian variogram models for field measured soil chemical properties. The ranges of spatial variability (A_0) exceeded the maximum sampling distance of 140 m for C/N, TC and TN, but were less for soil pH and EC. Except for soil pH, the $C/(C_o+C)$ values were close to unity for all chemical properties studied, implying highly developed spatial structure. The R^2 values were above 0.90 for all soil chemical properties, except for soil pH for which the R^2 was 0.87. Detrended residuals were fitted to the same spherical variogram models for pH and C/N ratio, but to linear for soil EC and TN and an exponential variogram for TC. The ranges of spatial dependence decreased for pH, TN and C/N, but increased for EC and TC. The R^2 values decreased for all soil chemical properties except for C/N. Overall, detrending soil chemical properties measured in 1999 resulted in a loss of spatial structure and a reduction in the

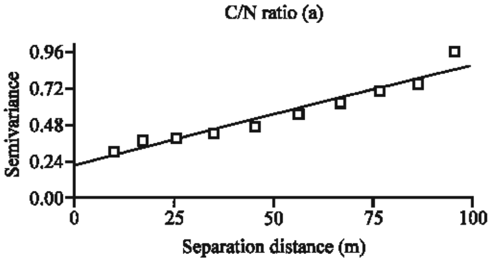


Spherical model (Co = 10.0000; Co + C = 4221.0000; Ao = 46.10; r^2 = 0.952; RSS = 748486)

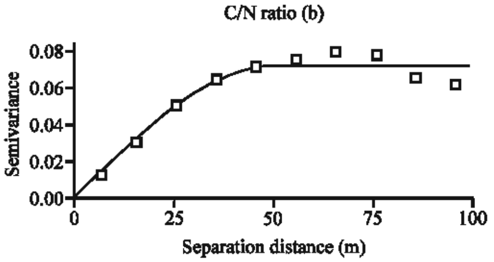


Linear model (Co = 2292.8798; Co + C = 2292.8798; Ao = 95.79; r^2 = 0.001; RSS = 6.703E+06)

Fig. 1b: Variograms of electrical conductivity (EC): (i) original data and (ii) de-trended residuals

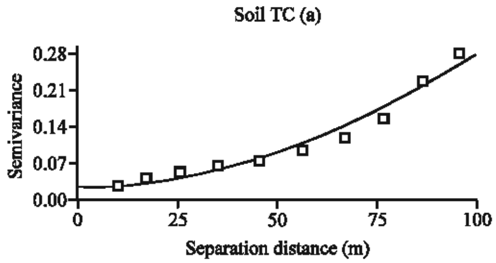


Spherical model (Co = 0.2130; Co + C = 1.6270; Ao = 310.90; r^2 = 0.929; RSS = 0.0264)

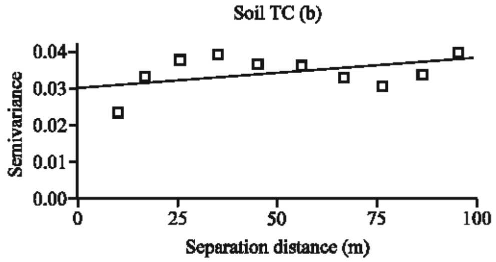


Spherical model (Co = 0.0001; Co + C = 0.0737; Ao = 50.80; r^2 = 0.941; RSS = 2.681E-04)

Fig. 1c: Variograms of carbon to nitrogen (C/N) ratio: (i) original data and (ii) de-trended residuals

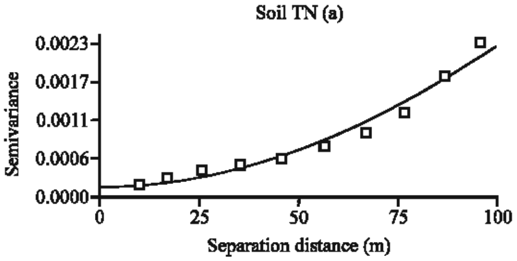

Soil TC (a)

Gaussian model (Co = 0.0230; Co + C = 1.9010;
Ao = 261.30; r^2 = 0.965; RSS = 2.241E-03)

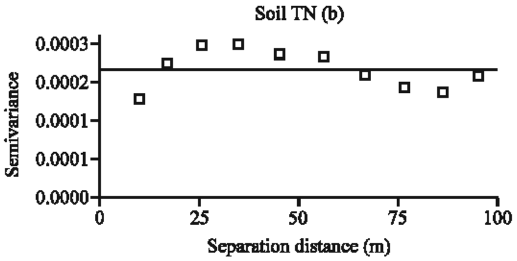

Soil TC (b)

Exponential model (Co = 0.0290; Co + C = 0.0580;
Ao = 310.90; r^2 = 0.119; RSS = 1.784E-04)

Fig. 2a: Variograms of soil total carbon (TC): (i) original data and (ii) de-trended residuals


Soil TN (a)

Gaussian model (Co = 0.0002; Co + C = 0.0155;
Ao = 260.80; r^2 = 0.967; RSS = 1.397E-07)


Soil TN (b)

Linear model (Co = 0.0002; Co + C = 0.0002;
Ao = 95.28; r^2 = 0.104; RSS = 9.616E-09)

Fig. 2b: Variograms of soil total nitrogen (TN): (i) original date and (ii) de-trended residuals

Table 5: Correlation between soil chemical properties and N_2O emissions in an onion field in Mikassa

	1999		2000			
	Ln (N_2O) (μg N_2O-N m^{-2} h^{-1})		N_2O (μg N_2O-N m^{-2} h^{-1})		Ln (N_2O) (μg N_2O-N m^{-2} h^{-1})	
Chemical properties	r	p	r	p	r	p
pH	-0.23	0.020	-	-	-	-
Log (pH)	-0.29	0.004	-	-	-	-
TN	-0.20	0.055	-	-	-	-
Log (EC)	0.25	0.012	-	-	-	-
NH_4^+	-	-	0.30	0.08	0.41	0.013
Log (NH_4^+)	-	-	-	-	0.43	0.009
Log (NO_3^-)	-	-	-	-	0.46	0.005
Log (NO_3^-+ NH_4^+)	-	-	-	-	0.48	0.003
NO_2^- (mg N kg^{-1} soil)	-	-	0.31	0.063	-	-
PO_4^{3-}	-	-	0.61	0.0001	0.46	0.005
Cl^-	-	-	-	-	-0.39	0.020

wellness of fit for EC, TC and TN, but an increase in both variogram parameters for C/N. Soil pH had a totally opposed behavior: an increase in spatial structure through detrending, but a reduction in wellness of fit. Soil chemical properties measured in 2000 were fitted to linear, exponential and spherical models. All ranges of spatial dependence were below the sampling distance. In addition, high R^2 were observed, except for nitrate (NO_3^-). These variograms fitted the data very well as symbolized $C/(C_o+C)$ values ranging from 0.87 to unity.

Mapping the Spatial Distribution of Soil Chemical Properties

Maps of soil pH, electrical conductivity (EC), carbon to nitrogen (C/N) ratio, total carbon (TC) and total nitrogen (TN) in an onion field in Mikassa in 1999, first degree trend surface and contoured residuals are shown in Fig. 3a-c and 4a, b. As for nitrous oxide emissions, estimation of soil pH, EC, C/N, TC and TN at unsampled locations was done using inverse distance weighing (IDW). A default grid spacing was used for interpolation purposes. After estimation of these soil chemical properties, isarithmic maps were produced with different contour levels. In 1999, maps showed that there is was systematic variability with spatial patterns for soil pH, EC, C/N, TC and TN in the onion field. However, maps produced after detrending data (contoured residuals) did not change these spatial patterns; either they remained the same (pH, EC) or increased in complexity (C/N, TN, TC).

Relationship between Nitrous Oxides Emissions and Soil Chemical Properties

The relationship between soil chemical properties and N_2O emissions in an onion field is shown in Table 5. In both 1999 and 2000, significant correlations were found between soil chemical properties and N_2O emissions, but only when the data was transformed into logarithmic scale. Several studies have also shown that it is difficult to directly correlate in situ N_2O emissions to soil mineral N contents and that N_2O emissions are related more to the soil N turnover rate than to mineral N pool size (Matson and Vitousek, 1990). It is also important to emphasize that all the N-radicals whether transformed or not were correlated with N_2O emissions in 1999 while they all correlated with N_2O emissions in 2000. The lack of consistency in the relationship between N_2O emissions and soil controlling factors has also been reported by several authors. In a study assessing the spatial variability of soil properties and gases emissions in southern Ohio, Jacinthe and Lal (2006) found stronger and statistically significant relationships between gas emissions and soil properties in July 2003 data. However, in May 2004, this trend changed and they observed ouly few statistical significant relationships. Jones *et al*. (2007) studied the influence of organic and mineral N fertilizer on N_2O

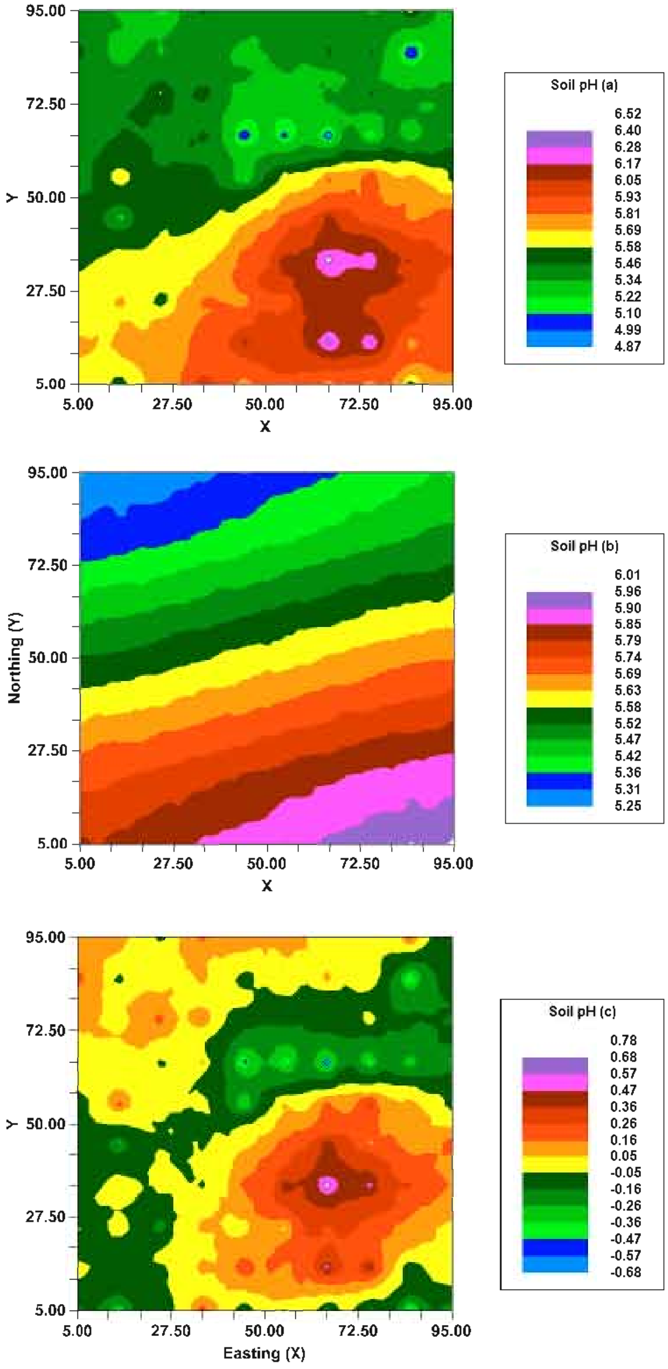


Fig. 3a: Maps of soil pH: (i) contour maps, (ii) first degree surface and (iii) contoured residuals

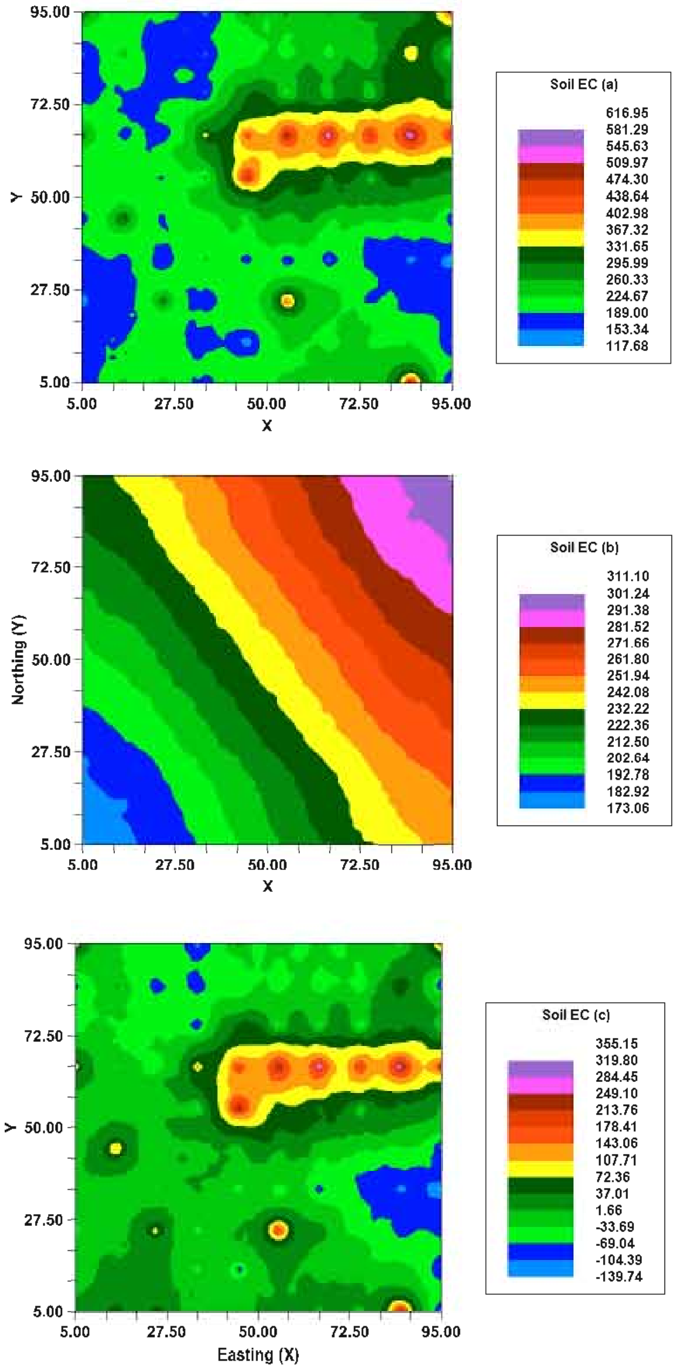


Fig. 3b: Maps of soil electrical conductivity (EC): (i) contour maps, (ii) first degree surface and (iii) contoured residuals

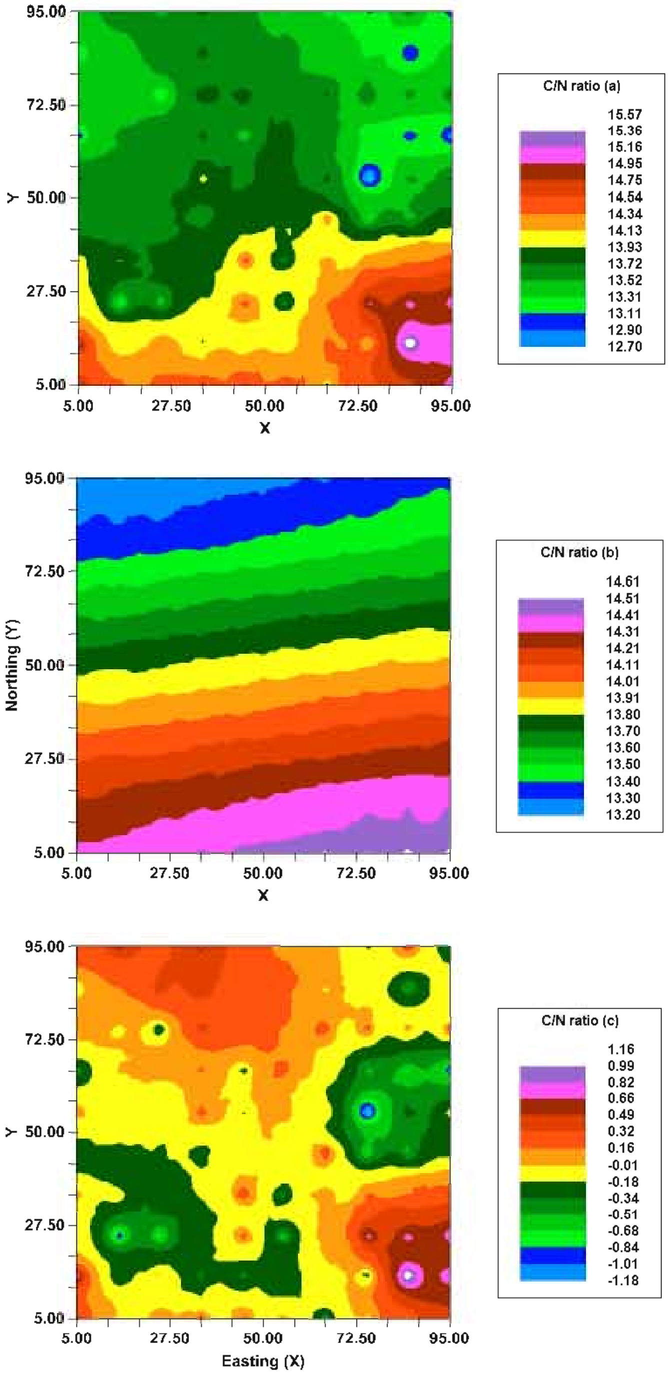


Fig. 3c: Maps of soil carbon to nitrogen ratio (C/N): (i) contour maps, (ii) first degree surface and (iii) contoured residuals

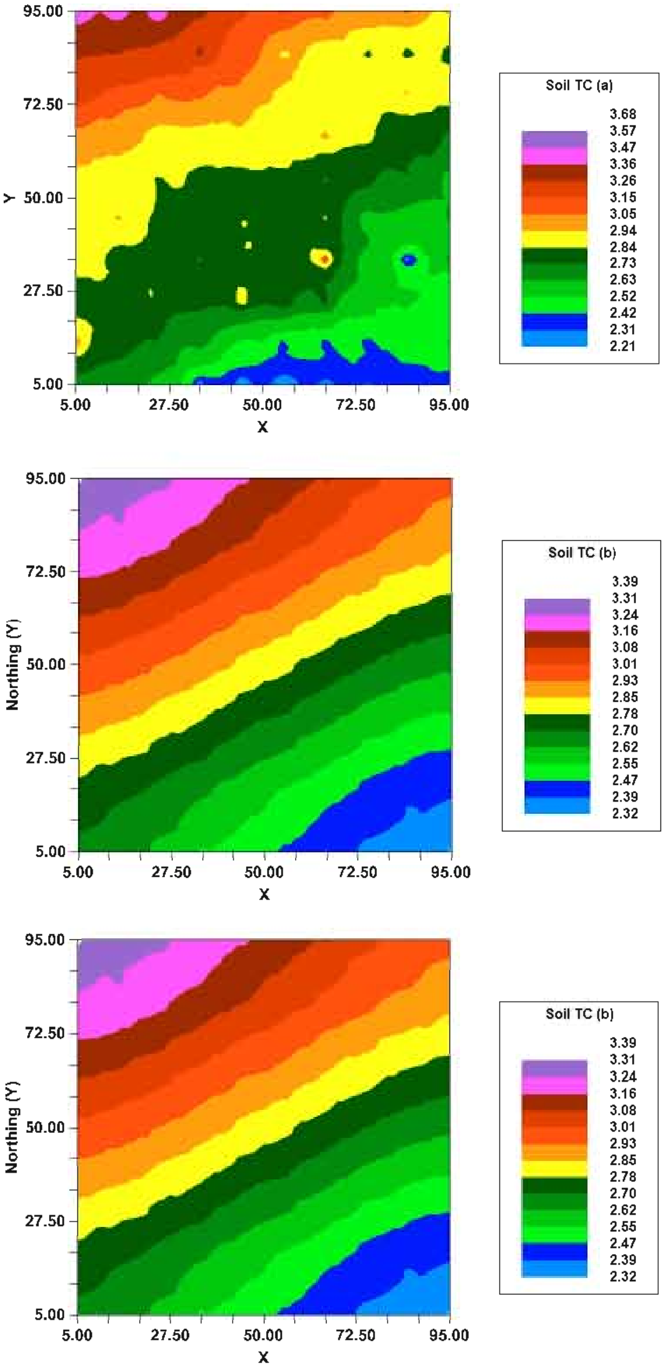


Fig. 4a: Maps of soil total carbon (TC): (i) contour maps, (ii) first degree surface and (iii) contoured residuals

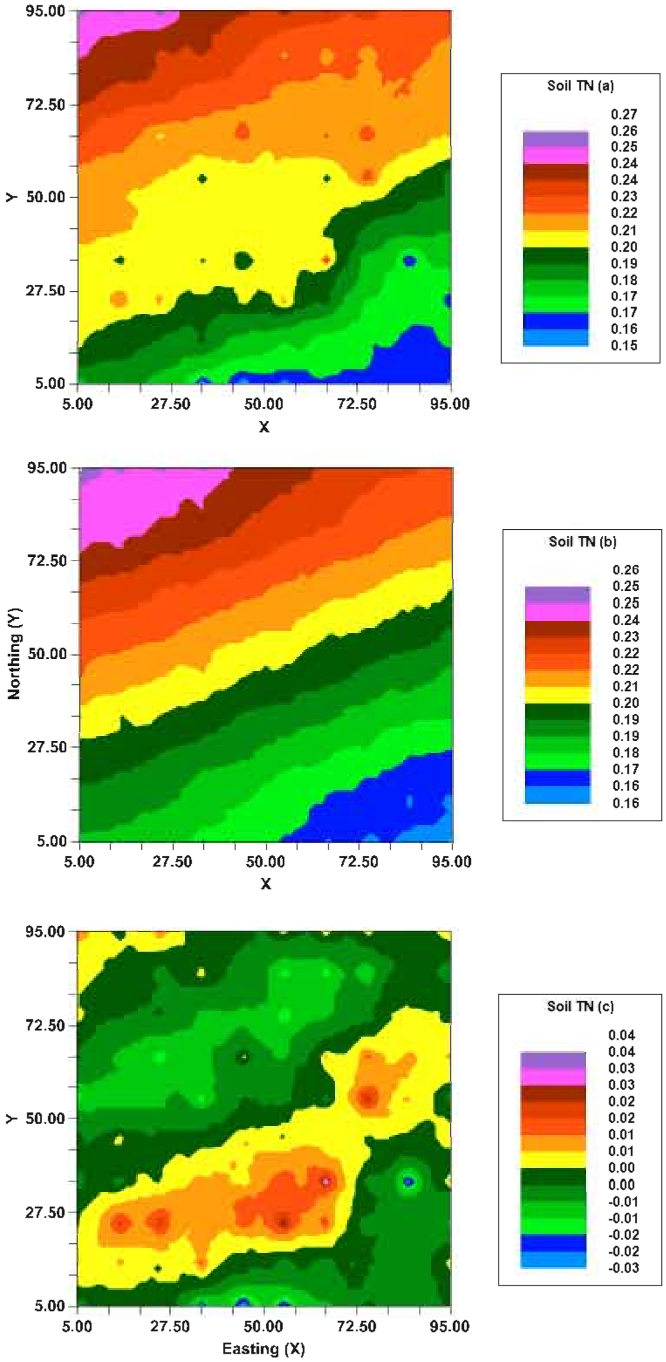


Fig. 4b: Maps of soil total nitrogen (TN): (i) contour maps, (ii) first degree surface and (iii) contoured residuals

emissions from a temperate grassland in Scotland. They investigated the influence of environmental conditions on N_2O emissions by a series of single and multiple regression analysis but could not find a relationship between N_2O emissions and several environmental factors. Paro *et al.* (2007) and Johnson *et al.* (2007) reported correlations between soil thermal properties and CO_2, CH_4 and N_2O emissions in Missouri forest and pasture, but they could relate soil chemical properties to these greenhouse gases. The most significant correlation between N-radicals and N_2O emissions was obtained with the snm of $NH_4^+ + NO_3^-$ ($r = 0.48$, $p = 0.0003$).

CONCLUSION

The objective of this study was to assess the spatial distribution soil chemical properties and relate these properties to N_2O emissions. The study was conducted in June 1999 and September 2000 in Mikassa, northern Hokkaido, Japan. This study confirms that fact N_2O emissions and soil chemical properties vary considerably in space and time, which make their studies more complicated. Variability of N_2O emissions and soil chemical properties 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. The study has also showed that there are other soil chemical properties that better correlate with N_2O emissions than nitrogen indices (NO_3^- and NH_4^+). Further investigations are therefore needed to study the relationship between N_2O emissions and soil chemical properties such as PO_4^{-3} which appear important soil controlling factors.

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