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Field Soil Properties Influencing the Variability of Denitrification Gas Fluxes

G. L. GRUNDMANN, D. E. ROLSTON,* AND R. G. KACHANOSKI

ABSTRACT

The spatial variability of field denitrification gas fluxes was investigated in relation to water content, soil-gas diffusivity, nitrate concentration, and water soluble organic C. Water was applied in a periodic fashion along a strip of Yolo soil (Typic Xerorthents) amended with chopped alfalfa hay and nitrate. Data were analyzed statistically using simple correlations and spectral and coherency analysis. Log spectrums showed that nitrate cycled at the frequency at which water content cycled and was negatively related to water content due to apparent leaching of nitrate from the high water application areas. Denitrification gas flux cycled at twice the frequency of water content with maximum fluxes occurring at the sides of the water peaks. Spectrums for water soluble organic C and gas diffusivity indicated no significant spatial cycling. Although denitrification gas flux was more highly correlated to soil-water content than to any other variable, coherency analyses revealed no significant relationships between denitrification gas flux and water or nitrate at specific frequencies due to opposing effects related to nitrate leaching and small gas diffusivities at the soil surface. The spatial pattern of denitrification calculated from a simple equation was examined using spectral analysis and was found to be not representing measured denitrification gas flux adequately at the frequency at which water content cycled. The discrepancy between measured denitrification gas flux and calculated denitrification rates was attributed to very small gas diffusivities preventing gas transport to the surface and inadequate characterization of actual C and nitrate concentrations at microsites.

EFFORTS AT UNDERSTANDING the global behavior of N_2O emissions from soils and research on increasing fertilizer N use efficiency have resulted in several studies on factors affecting denitrification in the field. It is well known that denitrification rates depend on oxygen (O_2), C, nitrate, pH, and other factors (Bremner and Shaw, 1958; Burford and Bremner, 1975; Parkin and Tiedje, 1984; Firestone, 1982; Bowman and Focht, 1974; Lalisse-Grundmann et al., 1983). Despite numerous laboratory studies, few field experiments have succeeded in significantly relating denitrification to the various soil factors known to affect denitrification (Burton and Beauchamp, 1985; Folorunso and Rolston, 1985; Colbourn et al., 1984; Ryden et al., 1979; Mosier et al., 1983). All studies determined very large spatial variability of gas fluxes in the field with coefficients of variation between 50 to 500% (Aulakh et al., 1982; Rice and Smith, 1982; Parkin et al., 1984; and Folorunso and Rolston, 1985). Large spatial variability in denitrification fluxes is probably related to a complex combination of the different factors each varying spatially and having individual effects on denitrification.

Recently, Folorunso and Rolston (1985), using a time series approach to determine if relationships could be established by evaluating data in the frequency domain, suggested that cycling detected in surface soil-water content was the cause of the cycling behavior observed in denitrification gas flux along a transect. The present research was undertaken to further develop insights into the causes of denitrification spatial variability by applying water in a periodic fashion along a transect, observing how denitrification and other soil parameters responded, and utilizing spectral and coherency time series analysis to evaluate the data.

MATERIALS AND METHODS

The data were derived from a field experiment conducted on Yolo loam, a deep well-drained alluvial soil, at Davis, California. A 45-m long strip, 2-m wide, was amended with finely chopped (<3 mm) alfalfa hay (6.7 Mg ha^{-1}) mixed in the top 0.1 m of soil. An irrigation system was then set to apply varying amounts of water to give soil-water contents which cycled along the transect. Three microsprinkler lines, with different patterns, were set in parallel but operated separately with line no. 1 run for 1 h, 3 d before the experiment. After 200 kg N ha^{-1} of $Ca(NO_3)_2$ was spread evenly, line no. 2 was run for 30 min. Then, 1 h before the experiment, the highest amount of water was applied with line no. 3 operating for 30 min. The distance between two maximum surface soil-water contents was 4.2 m.

Denitrification gas flux was measured every 0.7 m along the strip at 64 locations using the closed chamber method (Rolston, 1986) and the acetylene (C_2H_2) blockage approach (Ryden et al., 1979). Details are given by Grundmann and Rolston (1987). To obtain the denitrification flux measurements and the diffusivity in the shortest time and avoid changes in water content, the experiment was run in two separate halves on different days.

Within 1 h after flux measurements were made, gas diffusivities were measured using a modification of the method proposed by McIntyre and Philip (1964). One milliliter of Freon 13 was injected into the chambers used for denitrification flux measurements and the atmosphere was thoroughly mixed for 1.5 min using the fan. Diffusion of Freon into the soil was determined by sampling the chamber headspace at 15 and 30 min.

Immediately after gas flux and gas diffusivity measurements were completed, the soil inside the chamber was collected, separated into two layers 0 to 0.05 m and 0.05 to 0.1 m and each homogenized. The water content was measured gravimetrically. Water soluble organic C from a cold water extraction (Folorunso and Rolston, 1985) was analyzed on a Dohrmann CD-80 analyzer (Dohrmann, Santa Clara, CA), and nitrate-N determined on the same extract by a colorimetric procedure (Anderson, 1979).

The data were evaluated statistically using traditional correlation and spectral and coherency analyses (Robinson and Silvia, 1978). Spectral analysis is a statistical tool for transforming data from the time or spatial domain to the frequency domain by partitioning the total variance to various frequencies. Frequencies exhibiting large variances indicate a periodic or repetitive nature to the data at that particular frequency or period. The cospectrum is the breakdown of the covariance between variables as a function of frequency and is used to establish if relationships exist between vari-

G.L. Grundmann, Lab. de Biologie des Sols, UA 697, Univ. Claude Bernard Lyon I, 43 Bd 11 Novembre 1918, 69622 Villeurbanne, Cedex, France; D.E. Rolston, Land, Air and Water Resources, University of California, Davis, CA, 95616; and R.G. Kachanoski, Dep. of Land Resource Science, Univ. of Guelph, Guelph, Ontario, Canada N1G 2W1. Contribution from the Agric. Exp. Stn. and the Dep. of Land, Air and Water Resources, Univ. of California, Davis. Research supported partially by a scholarship from the Ministry of Industry and Research, Government of France and Charbonnage de France Chimie-Azote et Fertilisants, France. Received 25 Sept. 1987. *Corresponding author.

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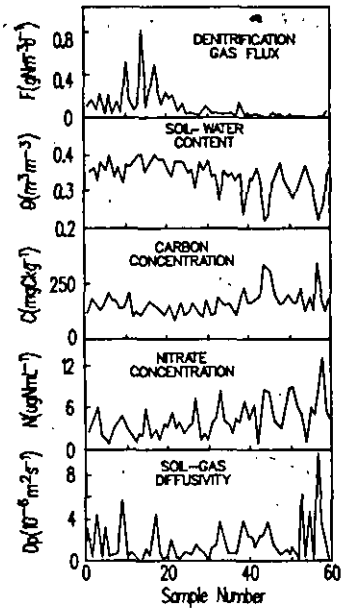

Fig. 1. Values of soil variables at 60 locations along the transect. Denitrification gas flux converted to rate per soil volume (0.1-m depth) in order to compare with calculated values.

ables at specific frequencies. The correlation between two series as a function of frequency can be calculated using the coherency spectrum (Brillinger, 1981) which is analogous to the r^2 value in ordinary regression analysis.

The choice of 60 measurements for spectral and coherency analysis was guided by the fact that water was applied to produce 10 periods, and 6 measurements were made within each period of 4.2 m (frequency = 0.167 cycles m^{-1}). When large trends in the data were detected, these series were detrended by subtracting the mean of each half of the data (for denitrification) or by subtracting the best fit linear or polynomial equation. Badly skewed data were transformed using the natural logarithm before running spectral or coherency analysis.

RESULTS AND DISCUSSION

Data for denitrification gas flux, water content, gas diffusivity, water soluble organic C, and nitrate along the transect are shown in Fig. 1. Denitrification gas flux dropped off drastically in the second half of the transect which was caused by reduced volumetric water content. The trend in water content is attributed to a similar trend of decreasing bulk density (not shown) for the second half of the transect. Nitrate concentration showed a tendency to increase slightly along the transect.

Table 1. Pertinent statistics for the various soil variables from the 0- to 0.1-m depth.

	Denitrification gas flux†	Water content	C concentration	Nitrate concentration	Gas diffusivity
	g N m^{-3} soil d^{-1}	m^3 H_2O m^{-3} soil	mg C kg^{-1} soil	μg N mL^{-1} soil solution	m^2 s^{-1}
Mean	0.10	0.35	47	427	1.7×10^{-6}
Variance	0.02	0.002	126	60 708	1.9×10^{-6}
Maximum	0.83	0.42	71	1 300	1.0×10^{-5}
Minimum	0.002	0.21	26	27	4.8×10^{-8}
CV	151%	13%	24%	58%	108%

† Surface flux converted to rate per soil volume (0.1-m depth) in order to compare with calculated values.

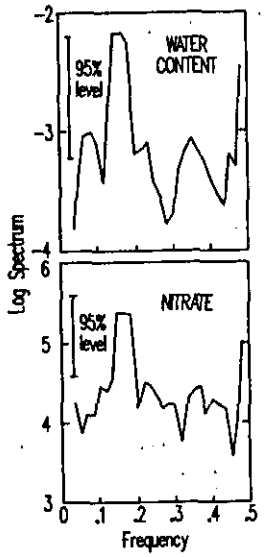

Fig. 2. Log spectrums of water content detrended according to a 3rd-order polynomial and nitrate concentration on a solution basis detrended according to a linear function.

Table 1 gives pertinent statistics for all 64 locations. As expected, denitrification flux varied more than the other variables. Table 2 gives mean values for the various soil variables if the total data set of 64 values is partitioned into subsets where denitrification was either > or < the mean of 0.1 g N m^{-3} soil d^{-1}, at the peaks of soil-water content, and at the peaks of denitrification flux. It is apparent from Table 2 that the mean soil-water content at the peaks of water content was only slightly larger than that at the peaks of denitrification flux. A change in mean water content from 0.38 to 0.33 was sufficient to cause denitrification flux to decrease to near zero values. The mean nitrate concentration was less at the peaks of water content than at other locations indicating that nitrate leaching likely occurred at locations of greatest water application. For the 0- to 0.05-m soil depth, the mean nitrate concentration at the peaks of water content was 134 μg N mL^{-1}. Water soluble organic C concentration was nearly constant for all subsets. The mean soil-gas diffusivity was slightly smaller at large denitrification fluxes and at peaks of water content than at other locations due to the effect of water content on the diffusivity.

Table 2. Mean values for the various soil parameters (0–0.1 m) partitioned into subsets according to level of denitrification flux and location of water content and denitrification flux peaks.†

	Denitrification gas flux	Water content	Nitrate concentration	WSC concentration	Gas diffusivity
	g N m^{-3} d^{-1}	m^3 m^{-3}	μg N mL^{-1}	mg C kg^{-1}	m^2 s^{-1}
All samples (64)	0.10	0.35	427	47	1.7×10^{-1}
Denitrification >0.1 (21)	0.25	0.38	303	49	1.0×10^{-6}
Denitrification <0.1 (43)	0.03	0.33	488	46	2.0×10^{-8}
Peaks of soil-water content (12)	0.15	0.38	239	49	1.2×10^{-6}
Peaks of denitrification (22)	0.19	0.37	337	49	1.1×10^{-6}

† Numbers in parentheses behind subset designations are the number of samples within that group.

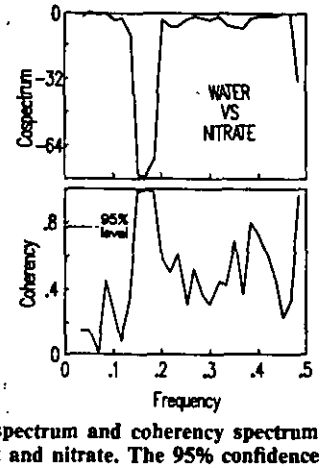

Fig. 3. The cospectrum and coherency spectrum between average water content and nitrate. The 95% confidence level for specific degrees of smoothing of the coherency spectrum is given on the figure.

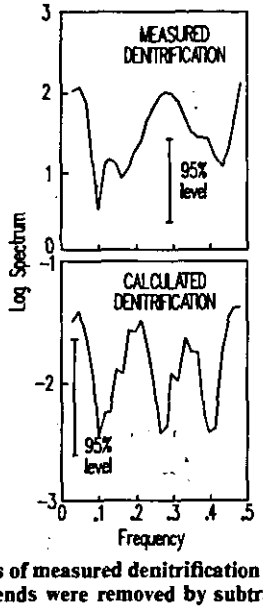

Fig. 4. Log spectrums of measured denitrification flux and calculated denitrification. Trends were removed by subtracting means from each half of transect.

Table 3 gives the correlation coefficients and confidence levels for selected pairs of soil variables (0–0.1m) if the total data set of 64 values is partitioned into subsets where denitrification flux was either high or low, at the peaks of soil-water content, and at the peaks of denitrification gas flux. For all data groupings, denitrification flux was more positively related to relative soil-water content (water content/saturated water content) than to any other variable, even at the peaks of water content where denitrification flux was less than at the peaks of denitrification flux. Although, these correlations between denitrification flux and water content explain no more than 58% of the variability, these correlations are larger than those observed in other field experiments (Burton and Beauchamp, 1985; Mosier et al. 1983; Folorunso and Rolston, 1985).

Nitrate concentration was negatively correlated to water content at a fairly high significance level at all locations except at the peaks of water content. Apparently, leaching of nitrate at the high water content locations resulted in poor correlation with soil-water content. Denitrification flux was negatively correlated with nitrate concentration for most data subsets due to the fact that water content and nitrate were negatively related.

Table 3. Correlation coefficient for several pairs of soil variables (0 to 0.1 m) partitioned into subsets according to level of denitrification flux and location of water content and denitrification flux peaks.†

	Denitrification vs. relative water content	Denitrification vs. nitrate conc.	Denitrification vs. C conc.	Denitrification vs. diffusivity	Nitrate vs. water content
All samples (64)	+0.56(99)	−0.33(99)	−0.07(41)	−0.22(92)	−0.83(99)
Denitrification >0.1 (21)	+0.49(98)	−0.19(59)	−0.31(83)	−0.06(20)	−0.45(96)
Denitrification <0.1 (43)	+0.52(99)	−0.43(99)	−0.07(35)	−0.26(91)	−0.84(99)
Peaks of soil-water content (12)	+0.76(99)	+0.20(48)	−0.11(27)	+0.30(66)	−0.20(47)
Peaks of denitrification (22)	+0.6899)	−0.32(86)	−0.36(90)	−0.09(30)	−0.68(99)

† Numbers in parentheses behind subset designations are the number of samples within that group. Confidence levels are given in parentheses within the table.

Denitrification flux was negatively related to gas diffusivity for all subsets except at the peaks of water content where a positive relationship existed. Although the correlation coefficient at the peaks of water content was small, this observation may indicate a limitation of denitrification gas flux due to small diffusion rates across the wet soil surface.

The correlation coefficient between gas diffusivity and soil-air content within the top 0.1 m of soil according to a power function relationship was only 0.59 which implies that other processes such as diffusion through cracks and other heterogeneities within the soil may also be complicating the transport of gas across the soil surface.

Figure 2 gives the log spectrums for average water content and nitrate concentration. At 0.17 cycles m^{-1}, which is the frequency of applied water, significant peaks in the log spectrum of the water content and nitrate series occurred. This verifies that the water content was indeed cycling at the frequency of applied water and that it also strongly affected the nitrate concentration. A cospectrum and a coherency spectrum of water content and nitrate concentration given by Fig. 3 shows a highly-correlated, negative relationship at the frequency of 0.17 due to apparent localized nitrate leaching. The average bulk nitrate concentration in the 0.10-m depth was probably not limiting for denitrifying activity, although the concentration was certainly low in the upper 0.05 m of soil and may have limited denitrification at the microsite level.

The spectrums of diffusivity and water soluble organic C (not shown) indicated no significant spatial cycling. The C values varied within a quite small range from 26 to 71 mg C kg^{-1} soil.

The log spectrum for measured denitrification flux (Fig. 4) showed that denitrification flux did not cycle at the frequency imposed by the water content (0.17) but at nearly double that frequency of 0.30. Inspection of the original data (Fig. 1) also shows that there were

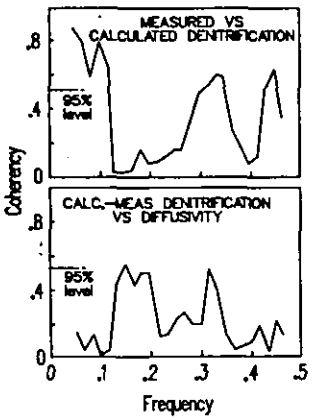

Fig. 5. Coherency spectrums for measured denitrification flux vs. calculated denitrification and calculated minus measured denitrification versus soil gas diffusivity. The 95% confidence levels are given on the figures.

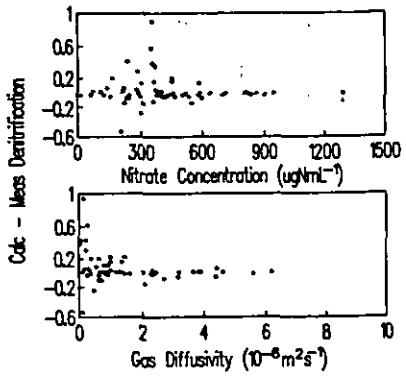

Fig. 6. Difference between calculated denitrification and measured fluxes as a function of soil gas diffusivity and nitrate concentration.

about twice as many peaks for denitrification flux as for soil-water content. Thus, the peaks of denitrification flux did not coincide with the highest water contents but occurred somewhere down the sides of the water content peaks to form, on average, two denitrification flux peaks for every water content peak. Since nitrate concentrations were lower at the peaks of water than other areas, this implies that a very narrow window of the combined effects of water content and nitrate concentration was required to produce the denitrification flux peaks. Since water content and nitrate did not cycle at the same frequency as denitrification flux, the coherency spectrums (not shown) between denitrification flux and water content and between denitrification flux and nitrate showed no significant correlation as a function of frequency.

Calculated denitrification was obtained using the equation proposed by Rolston et al. (1984)

$$F = kf_T f_w \theta C N \quad [1]$$

where F is denitrification rate, θ is volumetric soil-water content, N is NO_3^- concentration, C is water soluble organic C concentration, k is the denitrification rate constant, f_T is a temperature function set equal to 1, and f_w is a water function varying from 0 to 1 as an empirical means of accounting for anaerobic development in soil. The value of k used was that of Reddy et al. (1982). The water function was calculated using the equation proposed by Grundmann and Rolston (1987), $f_w = [\theta/\theta_s - 0.62)/0.38]^{1.74}$, where θ_s is the water content at saturation. Since f_w was developed from the same data set used in this paper, comparison of calculated denitrification from Eq. [1] with the measured flux is useful only in determining if Eq. [1] and a single function for f_w can reconstruct the pattern of spatial variability observed and to determine how Eq. [1] may be changed to better describe the denitrification process in the field.

The spectrum of calculated denitrification (Fig. 4) shows at least two peaks, one at the frequency of applied water (0.17), the other one at 0.32. The cycling of 0.32 corresponds to the cycling observed in the measured denitrification flux. The additional peak at 0.17 must be the result of cycling of water content. This peak is absent from the spectrum of measured denitrification flux, however, indicating that Eq. [1] is predicting peaks of denitrification in the high water content locations which are not occurring experimentally as peaks of denitrification gas surface flux.

The coherency between measured fluxes and calculated denitrification (Fig. 5) is high in the low frequency (large period) range and at the frequency where denitrification cycles with positive relationships for both frequency ranges. The high coherency at low frequencies (large periods) indicates that Eq. [1] does a reasonable job of predicting the large scale variation (overall high denitrification in the first half of the transect and low denitrification in the second half). The high coherency centered at the frequency of 0.32 is due to a good prediction of denitrification at the frequency at which measured denitrification flux cycled. The overall $r^2 = 0.36$ between measured flux and calculated denitrification, however, is fairly low due to low coherencies at other frequencies especially at the frequencies where the highest water contents occurred.

The coherency spectrum of calculated denitrification minus measured flux versus gas diffusivity (Fig. 5) shows two significant peaks at the frequencies where the water content cycled and at the frequency where denitrification flux cycled. The cospectrum (not shown) indicates a negative relationship for both peaks which indicates that as the diffusivity decreases, the difference between calculated denitrification and measured flux would increase. Evaluating the difference between calculated and measured denitrification as a function of diffusivity and nitrate content (Fig. 6) shows that the equation fails to predict the flux for very low diffusivities. These fluxes correspond to intermediate values of nitrate concentrations. Eq. [1] predicts denitrification rates greater than the measured surface flux for very high water contents because average nitrate concentrations in the bulk soil were substantially greater than those in microsites or that the transfer of N_2O gas to the surface may have been impeded by very low gas diffusivities. Both processes may explain why maximum denitrification surface fluxes did not occur at the maximum water contents.

Further evaluation of the difference between calculated denitrification and measured flux as a function of gas diffusivity (Fig. 6) shows that the largest discrepancies occurred at six locations where diffusivities were very small. All six data points of Fig. 6 with

positive differences between calculated denitrification and measured flux greater than 0.2 g N m^{-3} soil d^{-1} had diffusivities less than 3.3×10^{-7} m^2 s^{-1}. The next four largest positive differences have diffusivities less than 1.5×10^{-6} m^2 s^{-1}. This again suggests that very small values of gas diffusivity were partly responsible for the discrepancy between measured flux and calculated denitrification at the locations of greatest water content. If the 10 data points giving the largest positive difference between calculated and measured values are removed from both series, the correlation increases from 0.59 to 0.79, indicating that further efforts at increasing the capability of predicting the surface flux of denitrification gases should include a component accounting for gas diffusion to the soil surface.

SUMMARY AND CONCLUSIONS

This study demonstrates that denitrification gas flux did not cycle at the frequency of imposed soil-water content due to several interacting factors. Soil-water content variations were responsible for much of the variability of other parameters (especially nitrate) which subsequently affected denitrification flux through the development of anoxic sites, reduction in gas transfer to the soil surface by diffusion, and nitrate leaching. This interaction cannot be totally accounted for by the relationship proposed by Rolston et al. (1984).

Although relationships between the primary soil variables known to affect denitrification and field-measured denitrification gas fluxes exist, clear relationships are difficult to establish because of spatial interdependence of several of the variables, many of which have opposing effects upon denitrification. The simple model proposed by Rolston et al. (1984) helps to provide some predictive capability, but it still only explains about 37% of the observed variability. This model should be able to provide order of magnitude estimates of denitrification gas flux averaged over some area of soil but is not able to adequately make predictions of local variability based upon measured values of water content, nitrate, and C.

Both soil solution nitrate and water soluble organic C seem to be very poorly correlated to denitrification gas flux in the field. Thus, future research should be directed at improving the characterization of the C and nitrate pool available for denitrification in a dynamic field environment. The use of average values of C and N in the bulk soil is an oversimplification of actual substrate levels at sites of denitrification. Recently, Parkin (1987) has found that "hotspots" of high denitrification activity were associated with particulate organic C material, such as buried weed leaves, in the soil. A water soluble organic C extraction from bulk soil samples would not necessarily reflect the large C pool at such microsites.

Any improvement in predicting denitrification will also depend upon increased understanding and characterization of the locations of denitrification activity with soil depth, time, and spatial location. The results of this paper indicate that soil gas diffusion as related to both the development of anoxic sites and diffusive transport of product gases to the soil surface is an important process which should be considered if further predictive capability and understanding of the mechanisms causing the large spatial variability of denitrification gas fluxes are to be elucidated.

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