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Nitrate Distribution in a Deep, Alluvial Unsaturated Zone: Geologic Control vs. Fertilizer Management

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Introduction

For decades, nitrate leaching from agricultural sources (among others) has been a concern to agronomists, soil scientists, and hydrologists. Federal legislation first recognized the potential impacts to water resources in the early 1970s, when the Clean Water Act (CWA), the Safe Drinking Water Act (SDWA), the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA), and other water pollution related legislation was enacted. Since then, countless efforts have been mounted by both the scientific-technical community and the agricultural industry to better understand the role of agricultural practices in determining the fate of fertilizer and pesticides in watersheds (including groundwater) and to improve agricultural management accordingly.

From a groundwater perspective, much of the scientific work relating to nitrate has focused on two areas: documenting the extent of groundwater nitrate contamination; and investigating the fate of nitrogen in the soil root zone (including the potential for groundwater leaching) as it relates to particular agricultural crops and management practices. Rarely, these two research areas are linked within a single study and if they are, groundwater levels are typically close to the soil surface (less than 10 feet).

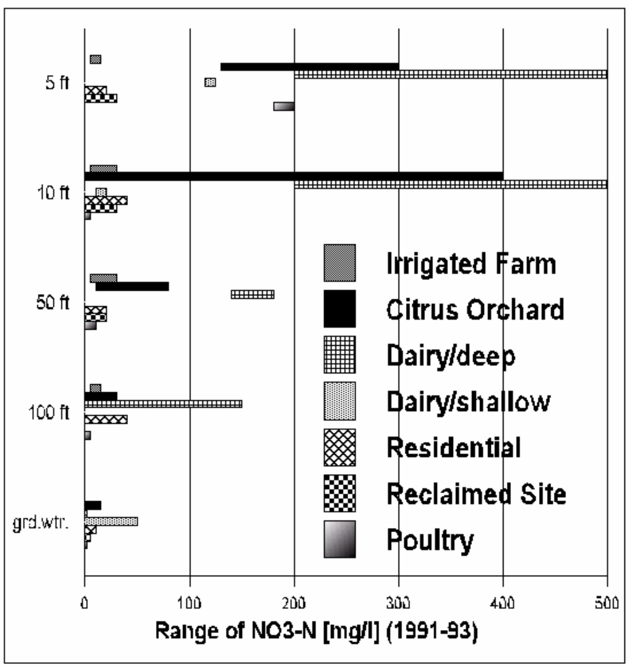

Figure 1: Range of nitrate-N concentrations at the bottom of the root zone and in the deep vadose zone. Water samples from the unsaturated zone and from groundwater were collected from seven instrumented access boreholes at seven sites in the Hemet basin, Southern California. Measurements were taken over a two year period (*data from*: Rees et al., 1995).

In California's valleys and basins, particularly in Central and Southern California, groundwater levels are frequently much deeper than 20 feet. The unsaturated zone between the land surface and the water table may therefore be from 20 to over 100 feet thick. Very few studies have investigated the fate or potential fate of nitrate in such deep unsaturated zones. Pioneering work on nitrate in deep soil profiles was presented by Pratt et al. (1972). They investigated nitrate profiles in a southern California citrus orchard to depths of 100 feet. The experimental site was subject to differential nitrogen treatment for 35 years from 1927 to 1962. Nitrogen treatments ranged from 50 to 350 lbs/ac. During the period from 1963 to the time of sampling in 1969, uniform treatment was applied at a rate of 150 lbs/ac. From their observations, the authors estimated that it would take between 10 and 50 years to leach nitrate to a depth of 100 feet.

Average nitrate-nitrogen levels below the root zone varied from 15 to 35 ppm under the 50 lbs/ac treatment and from 35 to 55 ppm under the 350 lbs/ac treatment. Based on gross mass balance estimates, denitrification at that site may have accounted for up to 50% of nitrate losses in the thick unsaturated zone profile where application rates were high. Lund et al. (1974) argued that nitrate losses (presumed to be due to denitrification) were strongly correlated with the textural properties of the soil. High losses were found in soils with pans or textural discontinuities, while losses were limited in relatively homogeneous, well draining soils. Later work by Gilliam et al. (1978), Klein and Bradford (1979), and Rees et al. (1995) in other areas of southern California supported these observations (Fig. 1), but provided little quantification of these losses.

We have recently initiated the development of a deep unsaturated zone hydrology research site, located in a former 'Fantasia' nectarine orchard at the Kearney Agricultural Center, Fresno County, California. The objective of our work is to provide a comprehensive assessment of the fate of nitrate in a 50 feet deep alluvial unsaturated zone that is not untypical of many agricultural areas in California. The site had been subject to differential nitrogen treatments during a 12-year experiment prior to an extensive drilling campaign in 1997. The assessment includes extensive geologic, hydraulic, as well as geochemical characterization. In this paper, we investigate the spatial distribution of nitrate in the deep vadose zone and analyze its relationship to the geologic framework of the site (intrinsic control) and the amount of nitrogen application (extrinsic control). Results have relevance with respect to the potential for denitrification before leaching soil water reaches the water table; and also with respect to designing a monitoring program of the deep vadose zone.

Methods

A 12-year fertilizer management experiment (from 1982 to 1995) was implemented in a nectarine orchard that consisted of 15 rows with 15 trees per row (Johnson et al., 1995). The orchard is located at the southern end of the research farm at the Kearney Agricultural Center near Parlier, Fresno County. Tree spacing within rows and between rows was 20 ft. The fertilization experiment consisted of five application treatments in a random block design with triple replicates. Treatments included nitrogen application rates ranging from 0 to 325 lbs/acre/year (not including nitrogen applied via irrigation water). Treatment plots consisted of five trees. Two border trees and one border row on either side separated treatments. For the subsurface characterization, three treatment plots were selected (0, 100, and 325 lbs/acre/year). In 1997, undisturbed soil cores were drilled with a direct-push drilling technique to a depth of 52 ft. At each of the three treatment plots, 18 cores were obtained (Fig. 2) and an additional six cores were drilled along a cross-section N-S through the entire orchard. A complete sedimentologic description by color, texture and moisture was made directly on the continuous core, prior to sample collection. 1,200 samples were collected (approximately one

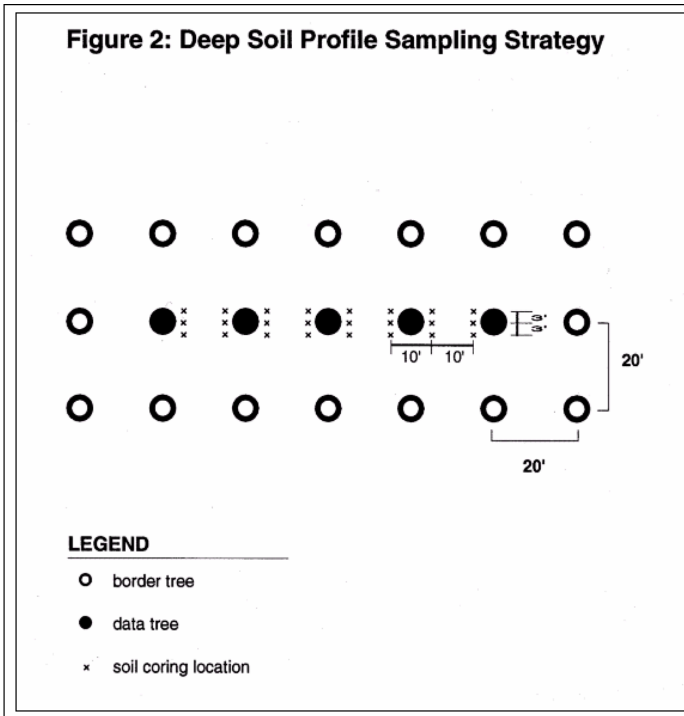

Figure 2: Location of the 52' deep unsaturated zone boreholes (marked by X) relative to the location of the tree rows (horizontal). One treatment shown only.

sample every 2.5 feet). Samples were collected for each sedimentologic stratum or sub-stratum. Soil (or more precisely: sediment) samples for nitrate analysis were two inches in length and 1.5 inches in diameter. Samples were preserved and stored for later analysis of physical and biogeochemical properties including soil texture, soil hydraulic properties (water content, unsaturated hydraulic conductivity and water retention functions; Tuli and Denton, 2001), and analysis of soil biochemical properties (pH, dissolved organic carbon, nitrate-N; Horwath & Paul, 1994). We are currently developing a protocol to also analyze for nitrogen and oxygen isotopic composition on low volume samples.

Results

Geologic Framework (Intrinsic Control)

The site is located on the Kings River alluvial fan, approximately 2 miles west of the current river channel. The alluvial unconsolidated sediments are derived exclusively from the hard, crystalline Sierran bedrock. They appear as intercalated, thick and thin lenses of clay, silt, sand, and gravel. The deposits contain fairly well sorted subangular to subrounded sand and gravel, and intercalated lenses of silt, sand and gravel with some lenses of clay, showing a downstream decrease in grain-size (Page and LeBlanc, 1969).

The material obtained in the borehole cores is exclusively composed of unconsolidated sediments. The top section of the core material is a recent soil (Hanford fine sandy loam). The sediments can be classified into textural groups ranging in grain-size from clay to pebble and cover a wide spectrum of silty and sandy sediments in between. The colors of the sediments range from grayish brown to yellowish brown, more randomly to strong brown (no significant reduction zones). The thickness of the beds varies from less than 1 cm for clayey material to more than 2.5 m for sandy deposits. Both, sharp and gradual vertical transitions are present between texturally different units. Five textural units are found the cores: 1) sand, 2) sandy loam, 3) silt loam/loam, 4) silt/clay loam/clayey silt/clay, 5) paleosol. The relative occurrence of each category in percent of the vertical profile length (in 5 cm sections) are 17.2% sand, 47.8% sandy loam, 13.8% silt loam/loam, 8.3% clay loam/clay and 12.9% paleosol.

The sand is quartz-rich, contains feldspar, muscovite, biotite, hornblende and lithic fragments consistent with the granitic Sierran source. Cross-bedding at the scale of few cm could be observed occasionally within fine-grained sand, showing reddish-brown layers intercalated with gray-brown ones. The dominant color of the sand is a light gray to light brown, the brown hue increasing with increasing loam content. The thickness of the sand beds is as much as 2.5 m and is dependent on the location of the core relative to the course of an ancient secondary distributary channel in which the sediments deposited. The channel appears to have a northeast-southwest orientation, diagonally through the orchard site. The mean thickness is 1.7 m. Very coarse sand and particles up to pebble grain-size (up to 1 cm) could be observed occasionally at the bottom of sand units, but were not present in all the cores. These are probably channel lag deposits and were laid down in deeper parts of the channels.

Sandy loam is the most frequent category within the profile. The color is usually light olive to yellowish brown. Some of the sandy loam sediments are considered to be weakly developed paleosols because of their stronger brownish color, root traces and presence of aggregates. Mean bed thickness is 50 cm. Individual beds can be as much as 2 m thick. The sorting is moderate to good. Clay flasers and thin (0.5-1 cm) clay layers occasionally occur in sandy loam units. Sandy loam sediments are assumed to have developed at the edge of channels, as levee or as proximal floodplain deposits near the channels.

Silt loam, loam and silty clay loam are usually slight olive brown to brownish gray in color. The bed thickness is within a scale of a few cm to dm. Fine grained sediments often show sharp contacts between the units. Changes from one unit to the next exist on small distances. Cross-bedding can more frequently be observed within silty sediments than in fine sands. Root traces and rusty brown colored spots are quite common. The depositional environment was presumably the proximal to distal floodplain of the alluvial fan, an area dissected by distributary branched braided streams.

The finest sediments are grouped in the 4th category: Silt, clay and clay loam. These are believed to have been deposited in the distal floodplain and in ponds that developed in abandoned channels. The main color is brownish gray to olive brown. Fine, less than 1 mm thick root traces and rusty brown spots are quite frequent also in the clay sediments. Statistics for the thickness of clay layers in the unit between 8 and 13 m depth show a mean thickness of 12.8 cm, but the mode is about 3 cm. A thick clay bed even extends to 50 cm and is observed in most of the cores.

Paleosols could be recognized in different stages of maturity. They show a brown to strong brown, slightly reddish color, exhibit aggregates, ferric nodules and concretions, few calcareous nodules and hard, cemented layers. They also display a sharp upper and a gradual lower boundary as is typical for paleosols (Retallack, 1990). Clay content decreases downwards in the paleosols. Another feature are fine root traces. Paleosols formed in periods of stasis marked by non-erosion and non-deposition, during the interglacials. Thickness of the paleosol horizons ranges from 50 cm to about 2 m.

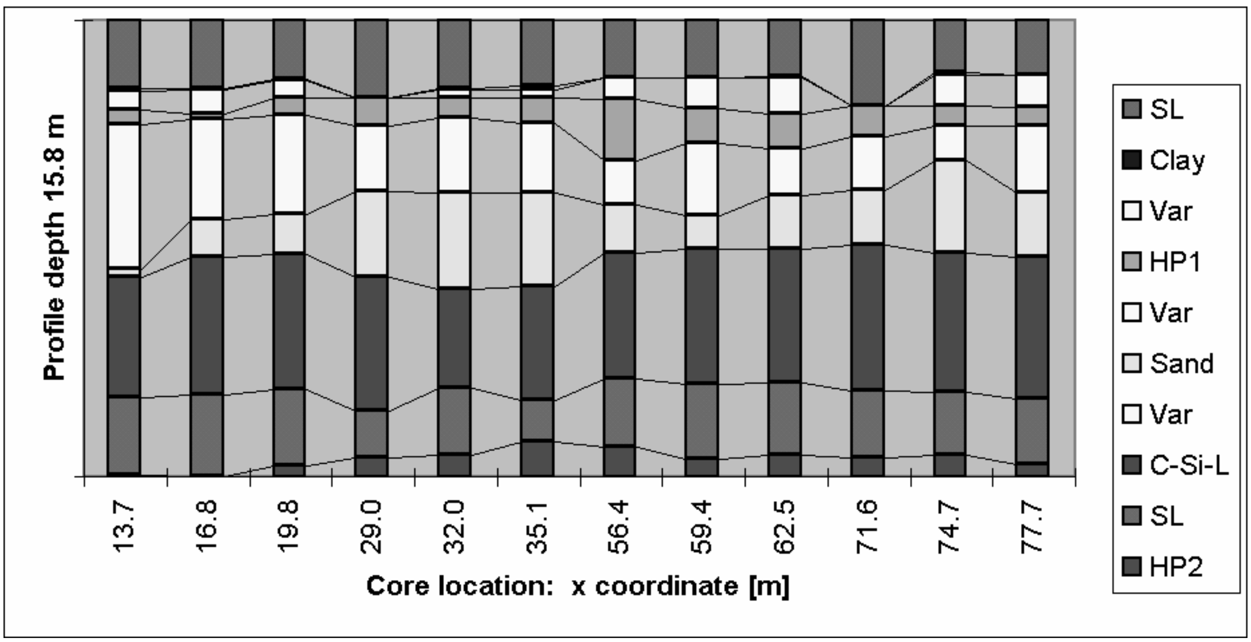

Figure 3: Stratigraphic cross-section along a tree-row showing the major stratigraphic units.

Several thicker units are recognized throughout the orchard and are used to construct a large scale geologic framework for the research site (Fig.3): The deepest parts of the cores from 15.8 to 15 m display a strong brownish colored, partly clayey paleosol hardpan. This paleosol marks the top of the Turlock Lake II formation (see below). From a depth of 15 to 12 m below surface, the main textural units are sandy loam to fine sandy loam, occasionally coarse sand and gravel, and occasionally fine-grained sediments right on top of the paleosol. In the cores with fine sediment at the bottom of this unit a coarsening-upward, in the other cores a fining-upward cycle can be observed. The sediments show a remarkable wetness due to proximity to aquifer water table. The sediments are vertically and laterally quite heterogeneous with relatively thin bedding (thickness cm to dm) between about 12 and 8 m depth, consisting mainly of clayey, silty and loamy material. Another strong brownish paleosol can be distinguished at a depth of 9-10 m. Between 9 and 6 m below surface a sand layer is found with laterally varying thickness averaging 1.7 m. A weak, mostly eroded paleosol is developed on top of the sand unit. Up to about 4-3 m below surface, sandy loam with intercalated sand, clayey and silty material is found. Different trends of upward-fining and -coarsening are found on top of each other and laterally next to each other within this unit. A 0.2 m to more than 1 m thick paleosol hardpan occurs at a depth of about 4-3 m. This paleosol marks the top of the Modesto formation. Sandy loam and subordinated loamy sand and loam are present from the top of the hardpan to the surface. 2.5 m below surface a laterally continuous clay horizon with a thickness of few cm is found in most of the cores.

Stratigraphically, the Quaternary deposits in this part of the valley can be divided into four units (Marchand and Allwardt, 1981). The Turlock Lake, Riverbank and Modesto Formations are of Pleistocene age (which began 2 million years ago). The Post-Modesto Formation belongs to the Holocene (which began 10,000 years ago). Most of the stratigraphic units found at the site are believed to represent separate alluvial episodes related to Sierran glaciations. The deposits are likely related to flood events that predominantly occurred during the end of a glaciation period. Paleosols, on the other hand, are indicative of substantial time intervals (several thousands to tens of thousands of years) between periods of aggradation (Marchand & Allwardt, 1981) and represent stratigraphic sequence boundaries. Paleosols are buried soil horizons that were formed on stable upper-fan, terrace or hillslope surfaces during interglacial periods (Lettis, 1982). At the site, they consist of strongly cemented sand to sandy loam with a characteristic reddish-brown color. Cementation is primarily by Fe-oxide and Mn-oxide, but also from calcification. They result from initial stratification or drainage boundaries in soil parent material (Harden & Marchand, 1977). Soils that formed on top of the upper Turlock Lake Formation are estimated to be 600 Ka (1Ka = 1000 years) old (Harden, 1987). The estimated age of the Riverbank formation is 130-450 Ka. The Modesto Formation corresponds to the most recent glaciation period (Huntington, 1980).

Nitrate Applications (Extrinsic Control)

Annual fertilizer applications in the three plots were 0 lbs/ac, 100 lbs/ac, and 325 lbs/ac. Granular N fertilizer was applied to the 14 - 16 feet wide, shallow broad furrows, but not to the center berm of the tree-row, which is 3-5 feet wide (tree spacing is 20 feet in either direction). The first 100 lbs were applied in the fall of each year using a tractor mounted spreader. Application uniformity was not measured, but anecdotal evidence indicates that higher amounts were applied near the edge of the furrows and less in the center of the furrows. In plots with N treatments above 100 lbs/ac, additional fertilizer was manually applied in the spring of each year. Application was limited to the area around individual trees (in a 3 x12 sq.ft. area in the furrows on either side of each tree). The orchard received further nitrogen from nitrate in precipitation (less than 5 lbs/ac) and from nitrate in irrigation water (30 - 50 lbs/ac assuming 4-5 mg/l of nitrate-N in 3-4 acft/ac of irrigation water). Nitrogen losses from the orchard are predominantly by fruit harvest. While crop yields varied little between treatmens, fruit N levels varied greatly from treatment to treatment. For the three treatments, harvest is estimated to remove 35, 70, and 110 lbs N /ac, respectively (*Scott Johnson, personal comm.*). Leaf N uptake and cover crop N uptake are assumed to be returned to the soil via leaf fall, decomposition, and mechanical incorporation into the soil. From an agronomic perspective, annual nitrogen leaching losses (either to leaching below the root zone or to denitrification) can be estimated based on a simple mass balance model for the root zone:

$$\text{net N Losses} = (\text{Fertilizer N} + \text{Irrigation water N}) - \text{Harvest N}$$

This simple approach neglects N volatilization during plant material and root decay. Based on this equation, net N losses are estimated to be on the order of 0 lbs/ac, 70 lbs/ac, and 250 lbs/ac, respectively. In the 0 lbs/ac plot, it is assumed that irrigation water N supplied the bulk of the nitrogen, while large lateral roots into neighboring tree-rows may have captured additional N. If all losses go to groundwater (no denitrification), at an annual net water leaching rate of approximately 2 acft/ac, the resulting concentration in the deep unsaturated zone leachate should be 0 mg/l, 10 mg/l, and 50 mg/l for the three plots, respectively.

Nitrogen Profile

Nitrogen profiles in individual boreholes are highly variable with little apparent correlation between adjacent boreholes (Fig. 4). Average nitrate concentration in deep soil water of the 0 lbs/ac and 100 lbs/ac treatment were 5 mg/l and 2 mg/l, respectively. Variability of nitrate concentrations in both plots is found to be very large, ranging from less than 1 mg/l in many samples to over 100 mg/l in a few soil samples. The 325 lbs/acre nitrogen treatment yielded the highest average nitrate-nitrogen concentrations in soil water (10 mg/l) due to a much larger number of samples with high N values: Almost a third of the nitrate-N samples exceeded concentrations of 10 mg/l (the maximum allowable groundwater quality limit). Much fewer samples than in the other two plots are found with nitrate-N levels less than1 mg/l. Despite the large variability, the nitrate application rate can be shown to have a statistically significant effect on mean nitrate levels (one-way analysis of variance at a p-level less than 0.05).

Nitrate samples were also grouped by depth, using the average thickness of the seven major stratigraphic units (Fig. 3) as an indicator. After log-transformation of the nitrate-N data (to account for their highly skewed distribution), statistically significant differences are found between mean nitrate levels in different stratigraphic units. However, depth-dependence is highly non-linear, that is, no general trend exists for either decreasing or increasing nitrate-N with depth. Depth-dependent mean N were grouped by treatment plots measure the interaction between the most prominent extrinsic control (fertilizer treatment) and the most prominent intrinsic control (geologic layering). Depth and treatment dependent nitrate-N mean (and confidence intervals) are shown in Fig.4. Analysis of variance at $p<0.05$ shows that the depth X treatment interaction is statistically significant, yielding distinctly different profiles at each treatment plot.

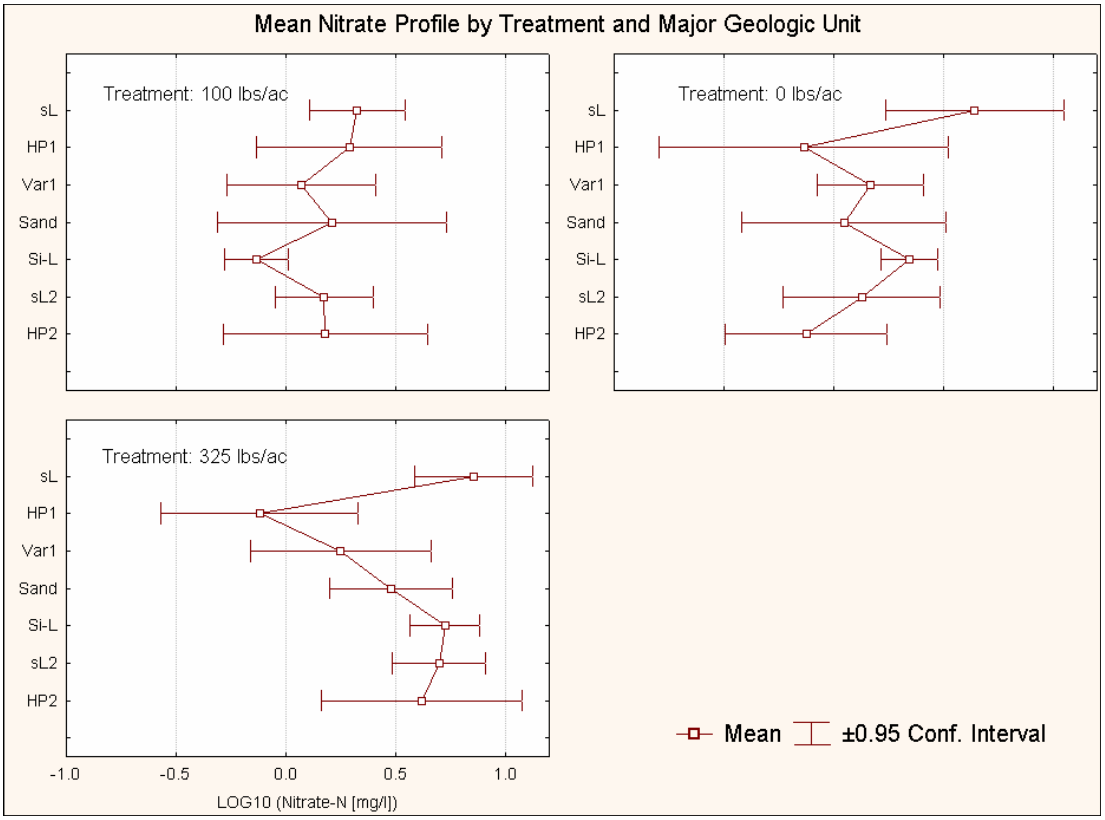

Fig. 4: Geometric mean nitrate profiles (arithmetic mean of the log10 of N) computed by taking group means for all possible treatment X depth (stratigraphic unit) combinations. Note that the x-axis shows Log10 of nitrate-N [mg/l], where -1 is equal to 0.1 mg/l, 0 is equal to 1 mg/l and 1 is equal to 10 mg/l.

Discussion

Given the small sample size, the large amount of spatial variability in the nitrate distribution is not surprising. Similar variability is found in other soil studies where soil samples have not been composited. The variability in nitrate levels is due to the high amount of spatial variability of both, the intrinsic and extrinsic controls. The fertilization treatment and the major geologic stratification depicted in Fig. 3 serve to explain only some of the observed variability in nitrate distribution. They only represent the most obvious spatial variability. Random within-treatment variability (random effects in extrinsic controls) and the high stratigraphic variability within each of the major geologic units further affects the nitrate distribution.

Extrinsic control. Random effects in the nitrogen loading distribution at the soil surface (extrinsic control) stem from the nonuniformity of the fertilizer application as described above. Limitations in the mechanical spreading and the intensional non-uniform distribution of the spring fertilizations immediately around the tree account for loading differences across spatial units that have length scale of one to several feet, but also at much smaller scales. Further random effects stem from the non-uniformity of the irrigation: at the largest scale, the top of a tree-row generally receives higher amounts of irrigation water than the bottom part of the tree-row. In the orchard, furrrow lengths were 300 feet and irrigation typically occurred over a 24 hour period. It is likely that the 0 lbs/ac and 325 lbs/ac treatments have received more irrigation water (and, hence, been subject to more nitrate leaching) than the 100 lbs/ac treatment, located within the bottom half of the orchard. This may partly explain why the 100 lbs/ac treatment has the least nitrate in the soil profile.

Laterally across furrows, irrigation uniformity was also limited due to undulations at the furrow surface. No nitrate loading occurred from the berms. However, no apparent differences are observed in the profile N concentrations in boreholes located under the center of the berms, when compared with those located in the furrow. The shallowest samples are taken at depths of 2 - 4 feet. The fact that these are not significantly nitrate-N depleted relative to the furrow boreholes indicates that lateral water movement redistributes nitrogen from the furrow into the berm area over a relatively short depth interval near the soil surface.

Random effects in unsaturated zone nitrate loading below the root zone may also be due to the non-uniformity of root nitrate uptake. Roots are clustered near the tree, although tree-roots may be several tens of feet long. These effects cannot be evaluated with our sampling scheme, since all boreholes are approximately the same distance from trees (~ 5 to 6.5 feet).

Finally, the top 10' (comprised primarily of fine sandy loam, sL) is considered to be also affected by the change in fertilization regime during the year prior to drilling: a single fall application of 100 lbs/ac occurred across all treatments after the original project was completed.

Intrinsic control. Effects of variability in geologic (intrinsic) control are more difficult to evaluate by statistical means alone. Uneven layering, small slopes of stratigraphic boundaries - even minor boundaries, and the random, intercalated occurrence of finer textured material are likely to lead to significant lateral (i.e., horizontal) water movement across distances of several inches to several feet (Harter et al., 1996). The lateral mixing of recharge water throughout the 50' unsaturated zone effectively disperses nitrate laterally across soil or sediment horizons. Within a few feet of the surface, such lateral water movement may completely mask the nitrogen variability imposed by spatially variable nitrogen loading at the surface. Over a depth of several tens of feet, lateral water (and nitrate) movement induced by the intrinsic heterogeneity of the unsaturated zone, may lead to a significant exchange of nitrogen even between treatment plots and neighboring tree rows, which receive a control application of 100 lbs/ac. If lateral mixing is significant, an increase of nitrogen with depth should be observed in the 0 lbs/ac treatment, whereas a decrease of nitrogen with depth should be observed in the 325 lbs/ac treatment (absent of any major other controls such as denitrification). While no such trend is apparent in the profiles of either the 0 lbs/ac or the 325 lbs/ac treatment, the overall nitrogen levels in the unsaturated zone differ much less from one another than would be expected based on the mass balance for each treatment: whereas mass balance predicted a difference in nitrate-N concentration of 50 mg/l, the actual difference is an order of magnitude smaller due primarily to much lower than expected N concentrations in the 325 lbs/ac treatment, but also due to higher than expected N concentrations in the 0 lbs/ac treatment. This is a strong indication for lateral exchange of nitrate between treatment and control plots.

The overall N loading from a field or orchard to groundwater, however, is not controlled by small scale variabilities in N application (random effects of extrinisic control). Generally, it is not even controlled much by the lateral movement of water within the unsaturated zone (random effects of intrinsic control) due to the limited extent of such movement relative to the size of a field. But it may be strongly controlled by the potential for denitrification between the root zone and the water table.

Intrinsic control of denitrification. Nitrate concentrations in the 325lbs/ac treatment are only 20% of the concentration expected from mass balance analysis indicating a significant potential for denitrification. However, a direct computation of the denitrification rate is not possible (without further modeling) due to lateral mixing effects and subsequent dilution underneath the research plots. The occurrence of denitrification is supported by preliminary isotope data.

On the other hand, if denitrification is significant, average nitrate concentrations should generally decrease with depth, particularly in the 325 lbs/ac and 100 lbs/ac treatment (no lateral inflow of higher nitrate levels from neighboring tree rows). But none of the three mean profiles show a monotonic decrease of nitrogen with depth. On the contrary, the 100 lbs/ac profile appears relatively uniform. The two other profiles have high concentrations in the top ten feet and very low nitrate concentrations in the upper hardpan at 10 - 12 feet (HP1). Below the hardpan N levels increase across the sand and into the silt-loams at depths of 30 - 40 feet. The nonuniformity of the mean profiles is surprising, given that the profiles are 50' deep, represent several years of consistent fertilization and water management and given that the signature of individual pulses of nitrogen applied at the surface are likely to dissipate within the top 10 to 15 feet due to dispersion. It is possible that the low nitrate samples from the upper hardpan represent stagnant pore water that was subject to temporary anaerobic conditions, particularly during the winter or during the irrigation season (ponding of water on top of the hardpan). However, the fact that nitrate concentrations are again higher below that hardpan indicates that significant amounts of nitrate-laden water pass through the hardpan quick enough to avoid denitrification.

Conclusions

1. Variability of nitrate concentrations throughout the unsaturated zone is extremely high, in part because of the small size of the samples, which were not composited.

2. The high variability underscores the importance of sampling from a large number of borehole samples at a given site. We question the significance of sampling only one or a few unsaturated zone boreholes to great depths. Results are uninterpretable with respect to the vertical distribution of nitrate or other solutes and have only limited statistical meaning with respect to the overall N content of the deep unsaturated zone. This finding has important consequences for monitoring of deep unsaturated zones: Since practically all standard observation tools of moisture and solutes in the unsaturated zone measure only small volumes (~ 1 liter or less), monitoring networks for individual sites must include multiple access holes ($\ggg 10$) to provide an adequate sample size.

3. We do not observe a strong stratigraphic control of denitrification with the exception of the hardpan, where significantly lower nitrate concentrations are observed in two of the three treatments. However, this may represent stagnant local water, since more mobile water that has moved below the hardpan has higher nitrate concentrations.

4. Preliminary isotope data and comparison of total N load in the deep unsaturated zone to that predicted from root zone mass balance suggest that some denitrification occurs, particularly where the N loss to below the root zone is relatively high.

To evaluate the effect of the spatially variable controls on N fate and transport in more detail, we are developing a deep unsaturated zone flow and transport supported by extensive hydraulic and geochemical field characterization. The model will allow us to evaluate effects of lateral water movement, superimposed by variability of both extrinsic and intrinsic controls on nitrate fate and transport. We hope to test several scenarios and hypotheses that have been raised by this analysis.

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