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Nitrogen Isotope Monitoring
at Unsewered Subdivisions

Nitrogen Isotope Monitoring at Unsewered Subdivisions

by

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Introduction

On-site septic-tank, soil absorption systems which serve unsewered subdivisions may cause nitrate-nitrogen levels in ground water to exceed the national drinking water standard of 10 mg/L of nitrate-nitrogen. Tinker, Jr. (1991 and 1990) analyzed water samples for nitrate-nitrogen from private water-supply wells in six subdivisions in Wisconsin. The nitrate-nitrogen values were analyzed for their distribution within the subdivision and for their relationship to the location of upgradient septic systems. In addition, Tinker, Jr. (1991) describes the application of a combined nitrogen mass balance model of Wehrmann (1984) and the BURBS nitrogen mass balance model of the Center of Environmental Research, Cornell University (1985) to the six subdivisions. The results of this investigation concluded that nitrogen from lawn fertilizer and septic-tank, soil absorption systems cause nitrate-nitrogen values to increase in the ground water beneath and on the downgradient side of the six subdivisions.

One other possible source of nitrate-nitrogen is agricultural fertilizer used on upgradient cultivated land. To evaluate this possibility, water samples from private water-supply wells in the six subdivisions in Wisconsin were tested for ^{15}N isotope. The assumption to the use of the ^{15}N isotope as an indicator of source of nitrate-nitrogen is that there is a lack of significant overlap of the range of ^{15}N isotope values for potential sources of nitrogen, and that isotopic fractionation does not occur to mask the original ^{15}N isotope signature of each source.

Natural Nitrogen Tracer Methodology

There are four radioactive and two stable isotopes of nitrogen. The radioactive isotopes have half lives which are too short for most uses, the half lives ranging from 0.11 seconds to 10.05 minutes. The stable isotopes ^{14}N and ^{15}N are commonly used as tracers. They occur naturally in a relatively fixed abundance with about 273 atoms of mass 14 for each atom of mass 15. The chemical properties of ^{14}N and ^{15}N are identical. The separation of the N isotopes depends on their differences in mass, and therefore, on physical properties (Hauck, 1973).

The natural variations in the N isotope abundance, designated as $\delta^{15}N$, are commonly expressed as parts per thousand differences from the ^{15}N to ^{14}N ratio in a standard, usually atmospheric N_2:

$$\delta^{15}N = \frac{(^{15}N/^{14}N)x - (^{15}N/^{14}N)atm}{(^{15}N/^{14}N)atm} \times 1000$$

A positive $\delta^{15}N$ indicates a higher ^{15}N concentration, and a negative value a lower ^{15}N concentration than in atmospheric N_2. The natural variations in N isotope ratios are small, usually within +10 isotope units, where one isotope unit is equal to about 0.000037 atom percent ^{15}N (Hauck, 1973).

Previous Studies

Natural variations of nitrogen isotopes have been used to identify sources of nitrate in surface water (Kohl et at., 1971) but Hauck et al. (1972), Edwards (1973), Bremmer and Tabatabai (1973) and Hauck (1973) questioned the method and interpretation of Kohl et al. (1971). Gormly and Spalding (1979) considered the above criticism valid as it applies to the study of Kohl et al. (1971). However, in Nebraska, Gormly and Spalding (1979) used natural variations of nitrogen isotopes as source indicators of animal waste versus agricultural fertilizer for areas of coarse-textured soils underlain by a highly permeable shallow vadose layer. Kreitler and Jones (1975) report the successful use of this technique to identify sources of nitrate in ground water in Texas, and Kreitler et al. (1978) report that differences in $\delta^{15}N$ with depth in the ground water of Long Island, New York were related to a change in land use. They suggested that the lighter $\delta^{15}N$ in the deeper aquifer (Magothy) was associated with recharge from cultivated fields during the agricultural period (pre-1950), while heavier $\delta^{15}N$ in the shallow aquifer reflected inputs from existing septic systems. Wolterink et al. (1979) collected over 300 soil and ground water samples for analysis of $^{15}N/^{14}N$ ratios from around the United States and representing a variety of environmental conditions. Standard statistical techniques were used to analyze the observed variations in $\delta^{15}N$ with respect to several nitrate sources and various environmental factors. It was concluded that nitrates from feedlots, barnyards, and septic tanks can be distinguished from natural soil nitrate on the basis of their $\delta^{15}N$. They cannot, however, be distinguished from each other. Spalding et al. (1982) suggest that low $\delta^{15}N$ in ground water in the Burbank-Wallula area of Washington indicate the primary source of nitrate contamination is from agricultural leachates. Flipse, Jr. and Bonner (1985) report on $\delta^{15}N$ from two heavily fertilized sites in Suffolk County, New York. The purpose of their study was to determine whether the $\delta^{15}N$ of fertilizer is increased during transit from land surface to ground water to an extent which would preclude use of this ratio to distinguish agricultural from animal sources of nitrate in ground water. They state that the nitrogen-isotope ratios of fertilizer-derived nitrate were not altered to an extent that would make them indistinguishable from animal-waste derived nitrates in ground water.

Study Sites

Four of the subdivisions are located in Eau Claire County and two in LaCrosse County in west-central Wisconsin (Figure 1 and Table 1). The subdivisions are situated on sand and loamy sand soil on glacial outwash or river terrace deposits. The soil slope is 0 to 6 percent in Oak Park and Pine Grove-Deer Park subdivisions and 0 to 2 percent in the other subdivisions. The mean depth to the water table, the mean depth of the water-supply wells, and the mean depth of the well casings for each subdivision are presented in Table 1.

All wells in Sandy Knolls, Oak Park, Briarwood and Lowes Creek subdivisions terminate in a water-table aquifer composed of sandy sediment. One exception is a well in Lowes Creek subdivision which terminates in sandstone under the sandy sediment. In Pine Grove-Deer Park subdivision, 21 wells terminate in the upper sandy sediment, 17 wells in sandstone under the sandy sediment, and one well in granite under the sandy sediment. The initial construction date, total area, number of septic systems, mean lot size, mean number of people per home, and mean number of bedrooms per home for each subdivision are presented in Table 2. The hydraulic gradient, hydraulic conductivity, average linear velocity of ground water, ground-water flow length across the subdivision, and travel time of ground water beneath each subdivision are presented in Table 3. The hydraulic gradients were determined from water-table maps constructed from data obtained from well construction reports of water-supply wells and elevations of permanent rivers. Hydraulic conductivity values were determined from specific capacity data obtained from well construction reports using the method described by Bradbury and Rothschild (1985). The average linear velocity of the ground water was calculated using Darcy's Law with porosity equal to 0.25. The distance across the subdivisions was obtained from aerial photographs, 7-1/2 minute topographic maps, or plat maps.

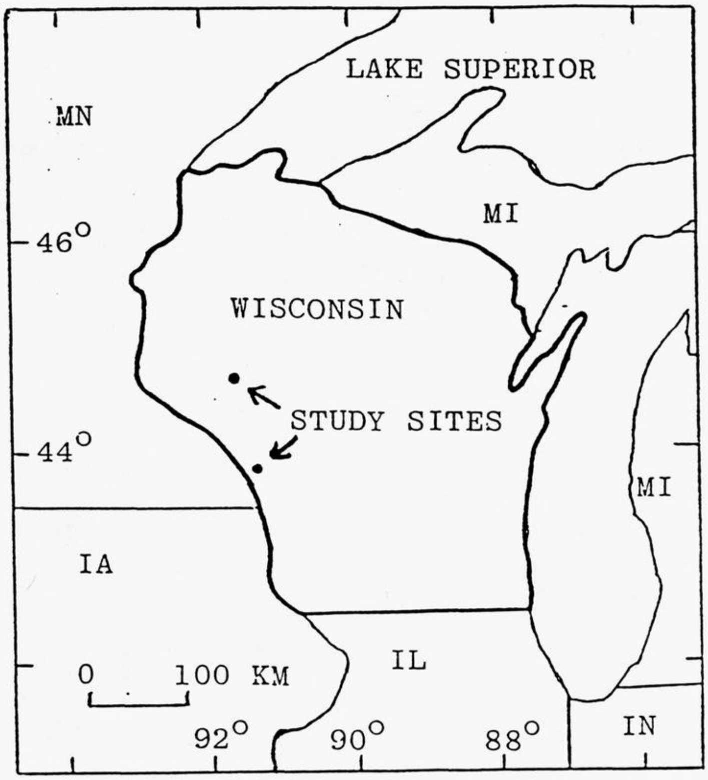

Figure 1. Location of study sites in Wisconsin.

TABLE 1. SITE LOCATION, SOIL, WATER TABLE, AND WELL CONSTRUCTION INFORMATION FOR THE SUBDIVISIONS.

Subdivision	Soils	Mean Depth to Water Table Feet	Mean Depth of Well Feet	Mean Depth of Well Casing Feet
Sandy Knolls NE 1/4, Sec 29, T17N, R7W	Sand Loamy Sand	64±3.3*	93±11.3	88±9.1
Pine Grove- Deer Park S 1/2, Sec 22, T27N, R8W	Sand Sandy Loam	30±8.4	61±17.4	53±13.9
Oak Park SE 1/4, Sec 29, SW 1/4, Sec 28, T17N, R8W	Sandy Loam, Loamy Sand	75±10.0	120±12.8	116±11.5
Lowes Creek SW 1/4, Sec 15, T26N, R9W	Loamy Sand	34±1.9	93±7.2	89±6.7
Briarwood S 1/2, Sec 22 T26N, R9W	Loamy Sand	33±6.5	74±15.8	62±21.0

* ± one standard deviation

TABLE 2. SITE CHARACTERISTICS OF THE SUBDIVISIONS

Subdivision	Initial Construction	Total Area Acres	No. of Septic Systems	Mean Lot Size Acres	Mean No. of People Per Home	Mean No. Bedrooms Per Home
Sandy Knolls	1981	39	50	0.5	3.9	3.4
Pine Grove-Deer Park	1971	104	70	1.1	3.9	3.1
Oak Park	1972	89	128	0.6	4.0	3.1
Lowes Creek	1975	55	33	1.3	3.3	3.4
Briarwood	1978	81	45	1.2	3.2	2.9
Mill Run (Tinker, 1987)	1974	72	69 Duplexes 18 Singles	0.5	3.0	---

TABLE 3. HYDROGEOLOGICAL CHARACTERISTICS OF THE SUBDIVISIONS

Subdivision	Hydraulic Gradient	Hydraulic Conductivity Feet/Sec	Average Linear Velocity of Ground Water Feet/Sec	Flow Length Feet	Travel Time Years
Sandy Knolls	.0026	$5.9\text{X}10^{-4}$	$6.14\text{X}10^{-6}$	1600	8.3
Pine Grove-Deer Park	.0091	$3.30\text{X}10^{-4}$	$1.20\text{X}10^{-5}$	1100	2.9
Oak Park	.0026	$1.10\text{X}10^{-3}$	$1.14\text{X}10^{-5}$	1400	3.9
Lowes Creek	.0110	$5.28\text{X}10^{-4}$	$2.32\text{X}10^{-5}$	1300	1.8
Briarwood	.0074	$4.63\text{X}10^{-4}$	$1.37\text{X}10^{-5}$	2000	4.6
Mill Run	.00033	$1.30\text{X}10^{-3}$	$1.71\text{X}10^{-6}$	1400	26.0

To reduce ambiguity in the analysis, five of the six subdivisions selected are located in areas with little to no upgradient agricultural activity or other known sources of nitrate-nitrogen.

Sampling Procedures of Water Supply Wells

Water samples were collected from inside or outside faucets of homes in each of the six subdivisions. The samples were analyzed for nitrate-nitrogen at the Wisconsin State Laboratory of Hygiene in Madison, Wisconsin. The samples were analyzed for $\delta^{15}N$ by Roy Spalding, Ph.D. at the Water Center, 103 Natural Resources Hall, University of Nebraska, Lincoln, NE. The University of Nebraska laboratory is certified by the U.S. Environmental Protection Agency to conduct ^{15}N analyses.

The wells selected for sampling had previously recorded values of nitrate-nitrogen in excess of 10 mg/L or were wells of highest nitrate-nitrogen within the subdivision. A larger number of samples were collected in Sandy Knolls, Oak Park, and Mill Run subdivisions where a larger number of private-water supply wells exceed 10 mg/L nitrate-nitrogen. Water samples containing 10 mg/L or greater of nitrate-nitrogen were requested by Roy Spalding of the University of Nebraska.

Water samples were collected following standard procedures. For example, nitrate-nitrogen samples were collected from taps after flushing, collected prior to any treatment system, placed in insulated and cooled containers, and shipped to the laboratory on the day of collection. For $\delta^{15}N$ samples, one liter of water was collected, frozen, and shipped in styrofoam containers the day of collection. A duplicate $\delta^{15}N$ sample showed a variation of 1.1.

Results

Thirty-eight samples were collected for $\delta^{15}N$ and nitrate-nitrogen analyses: two in Lowes Creek subdivision (Figures 2 and 3), nine in Oak Park subdivision (Figures 4 and 5), five in Pine Grove-Deer Park subdivision (Figures 6 and 7), three in Briarwood subdivision (Figures 8 and 9), eight in Sandy Knolls subdivision (Figures 10 and 11), and eleven in Mill Run Subdivision (Figures 12 and 13). Table 4 presents the mean and standard deviation of the $\delta^{15}N$ and nitrate-nitrogen values for each subdivision. The mean $\delta^{15}N$ for all measurements is 6.2 ± 1.6 and the mean NO_3-N value for all measurements is 10.6 mg/L $\pm$ 4.3 mg/L. The correlation coefficient between $\delta^{15}N$ versus nitrate-nitrogen for each well is 0.10 which is <90% significant by the t-Test (Davis, 1986).

Discussion

Figure 14 shows the range of $\delta^{15}N$ for each subdivision; the range of $\delta^{15}N$ presented by Gormly and Spalding (1979) for fertilizer N, fertilized soil NO_3-N, soil organic N, unfertilized soil NO_3-N, and animal waste N; the average $\delta^{15}N$ (10.9) plus and minus one standard deviation for septic tank waste and the average $\delta^{15}N$ (12.4) plus and minus one standard deviation for animal waste as presented by Wolterink et al. (1979); the range of $\delta^{15}N$ for fertilizer, cultivated land, unfertilized land, and animal waste in West Texas as presented by Kreitler (1979); the average $\delta^{15}N$ (0.2) for fertilizer used at a potato farm on Long Island, New York and the average $\delta^{15}N$ (6.2) for ground water beneath the potato farm as presented by Flipse, Jr. and Bonner (1985); and the average $\delta^{15}N$ (-5.9) for fertilizer used at a golf course on Long Island, New York and the average $\delta^{15}N$ value (6.5) for ground water beneath the golf course as presented by Flipse, Jr. and Bonner (1985).

NITROGEN-15 ISOTOPES (‰) LOWES CREEK 3/90

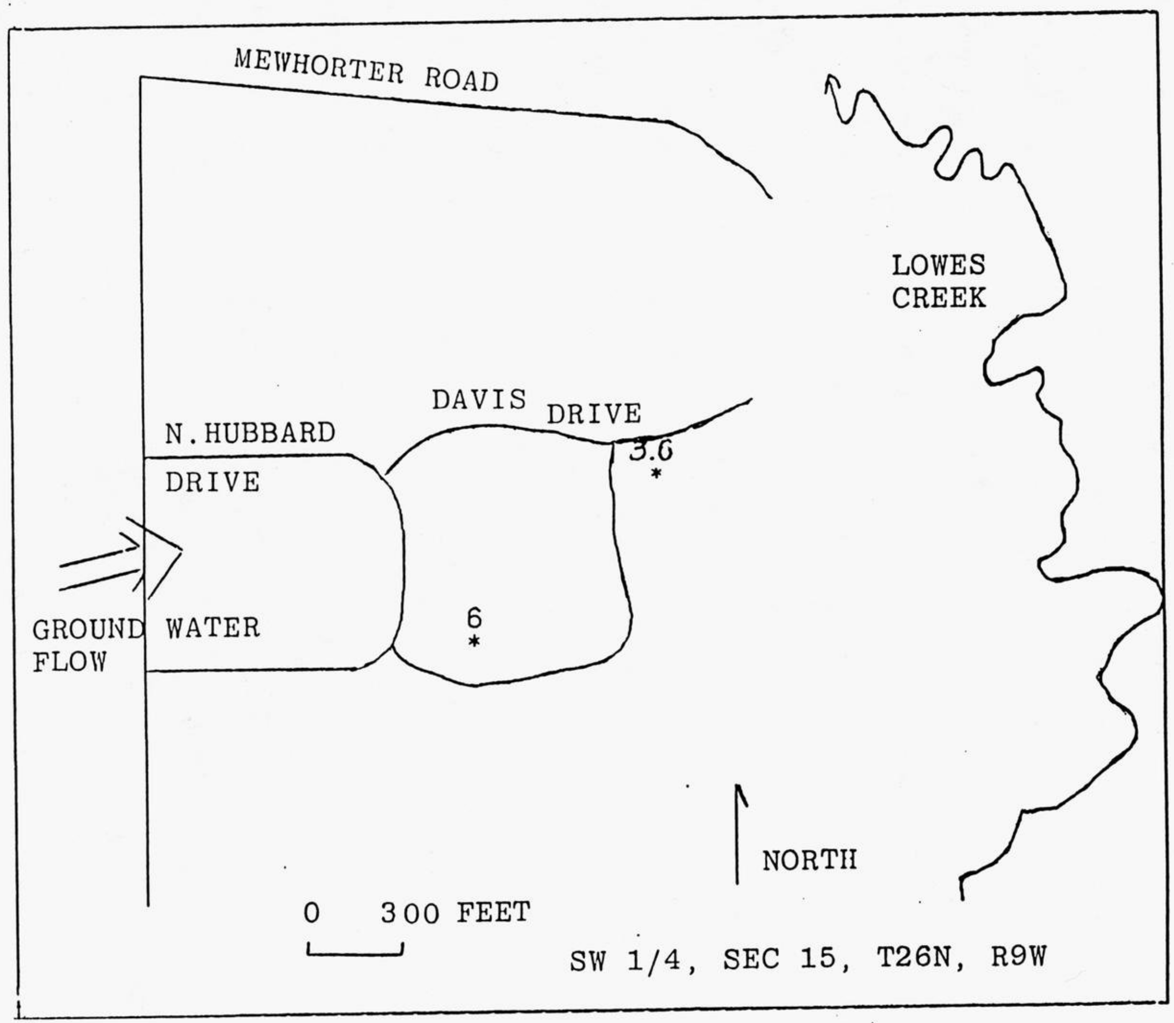

Figure 2. Nitrogen isotope values in parts per 10^3 differences, $\delta^{15}N$ for private water-supply wells in Lowes Creek subdivision, March 1990.

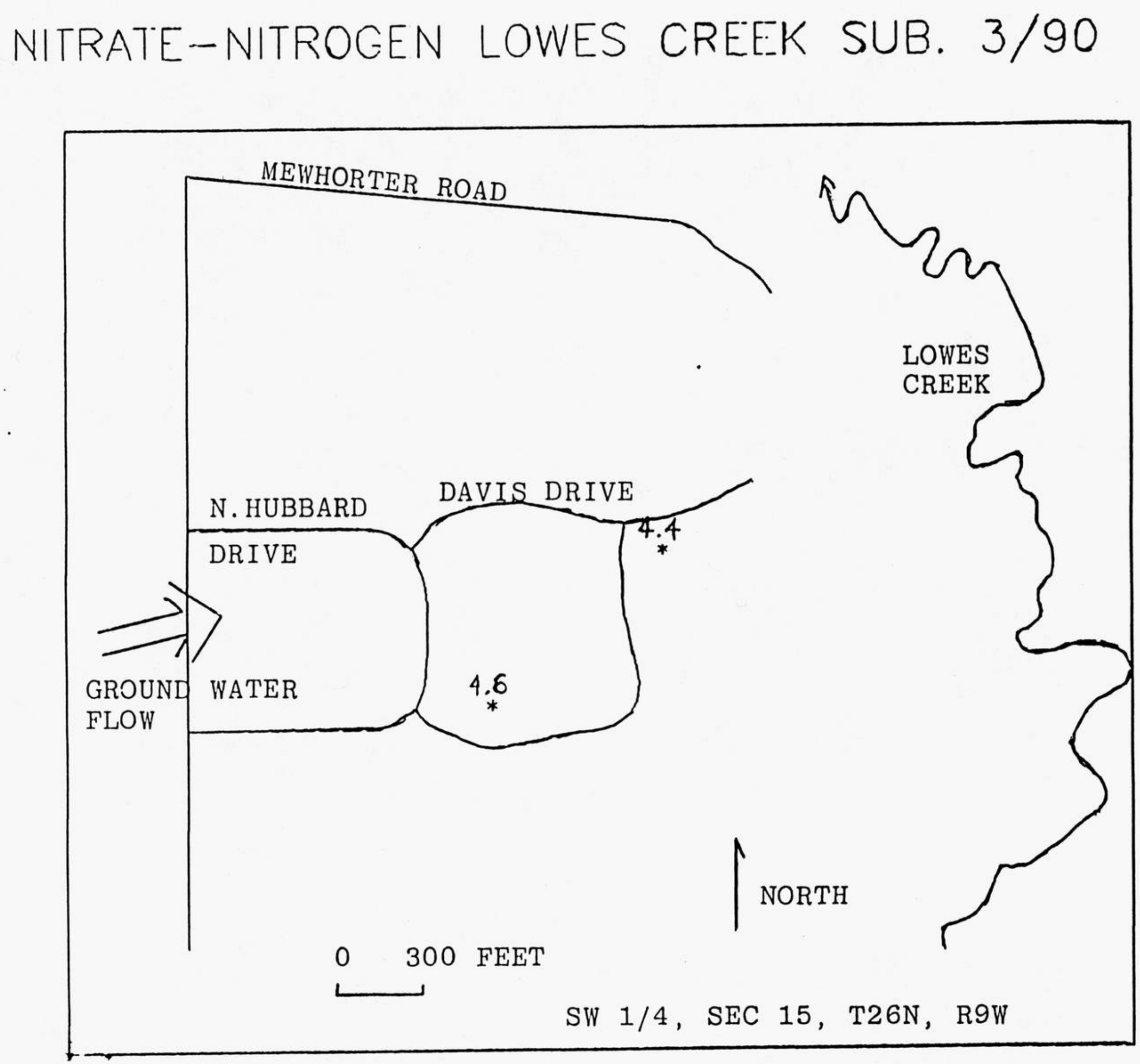

Figure 3. Nitrate-nitrogen in mg/L for private water-supply wells in Lowes Creek subdivision, March 1990.

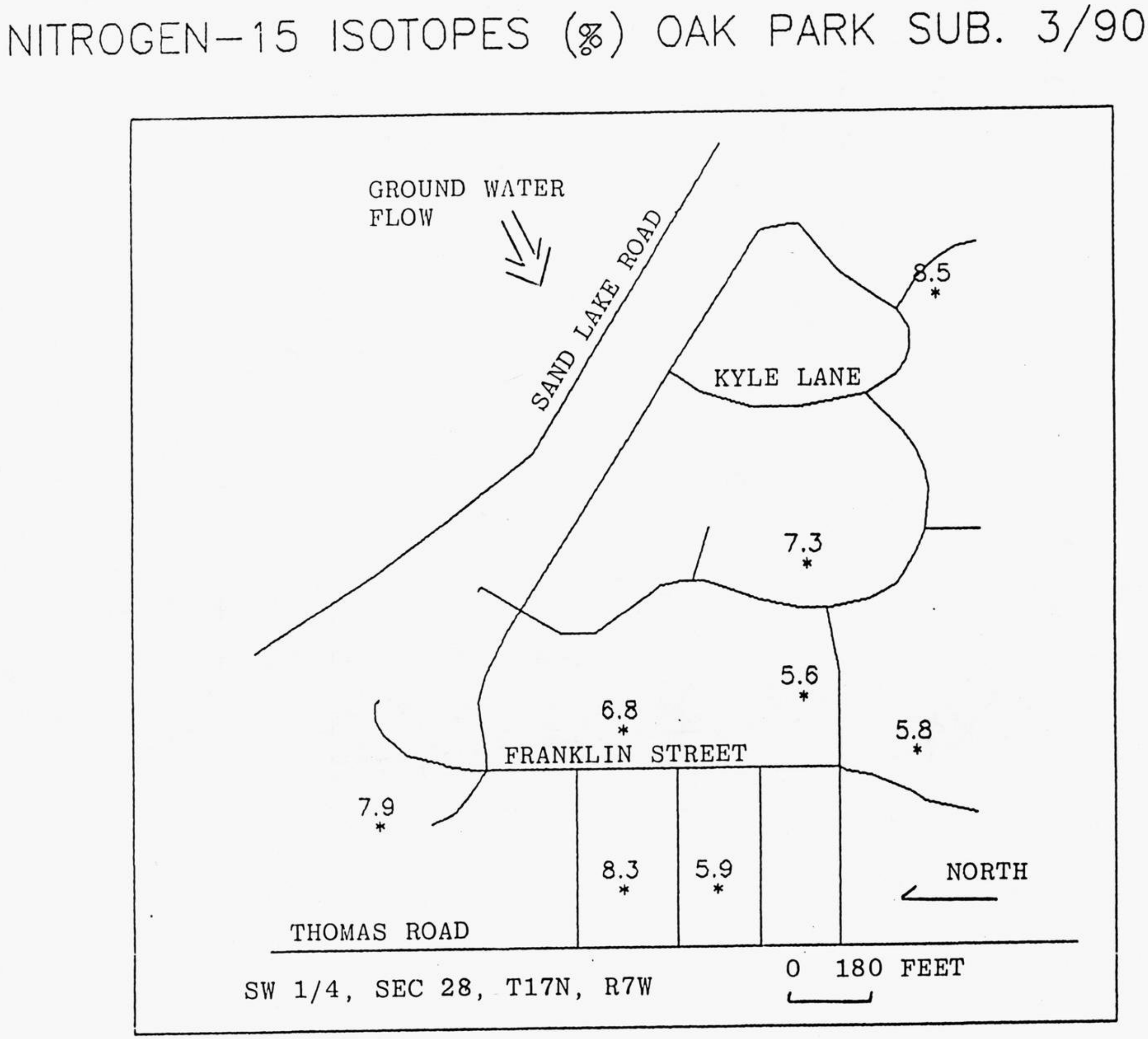

Figure 4. Nitrogen isotope values in parts per 10^3 differences, $\delta^{15}N$ for private water-supply wells in Oak Park subdivision, March 1990.

NITRATE–NITROGEN OAK PARK SUBDIVISION 3/90

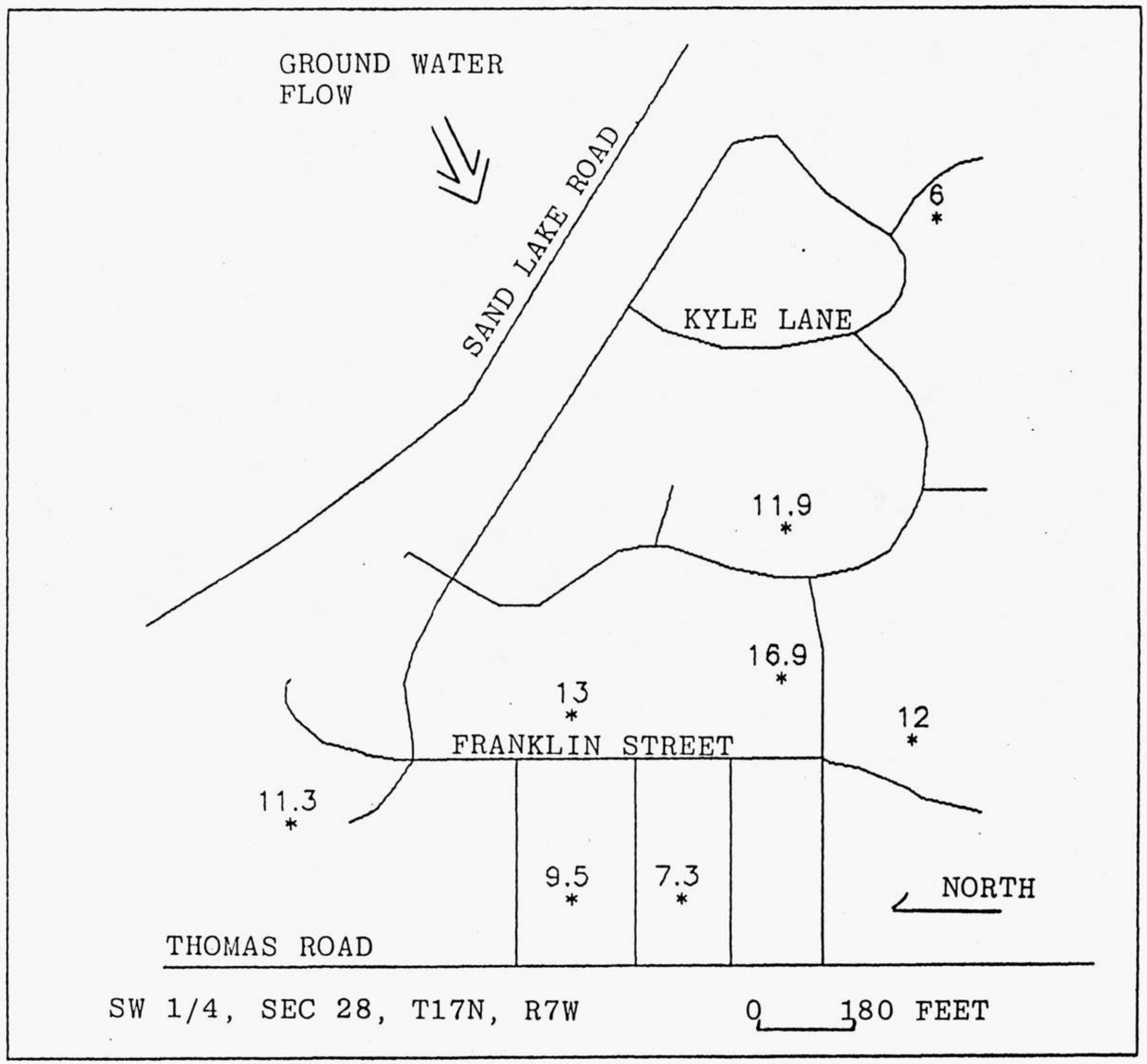

Figure 5. Nitrate-nitrogen in mg/L for private water-supply wells in Oak Park subdivision, March 1990.

NITROGEN-15 ISOTOPES (‰) PINE GROVE 3/90/90

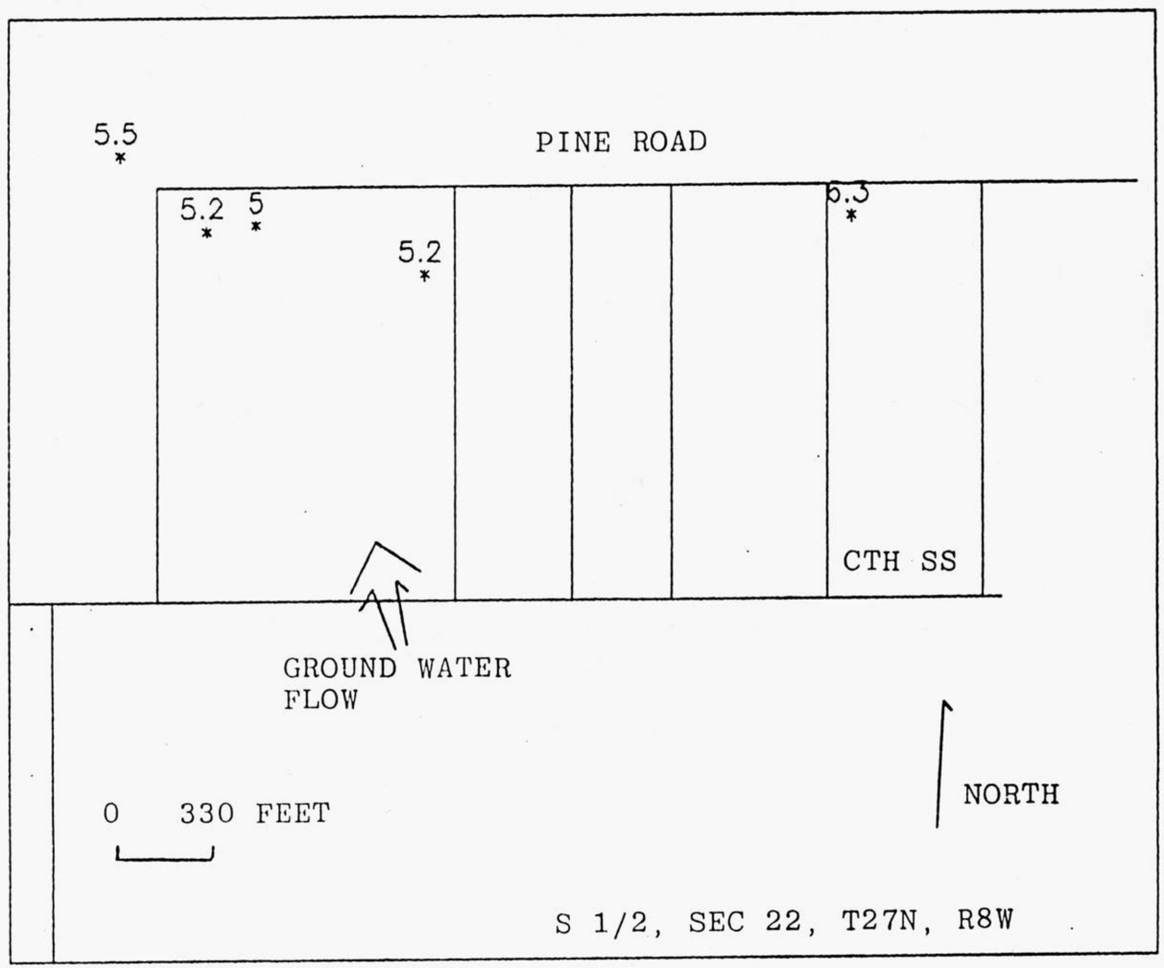

Figure 6. Nitrogen isotope values in parts per 10^3 differences, $\delta^{15}N$ for private water-supply wells in Pine Grove-Deer Park subdivision, March 1990.

NITRATE-NITROGEN PINE GROVE-DEER PARK 3/90

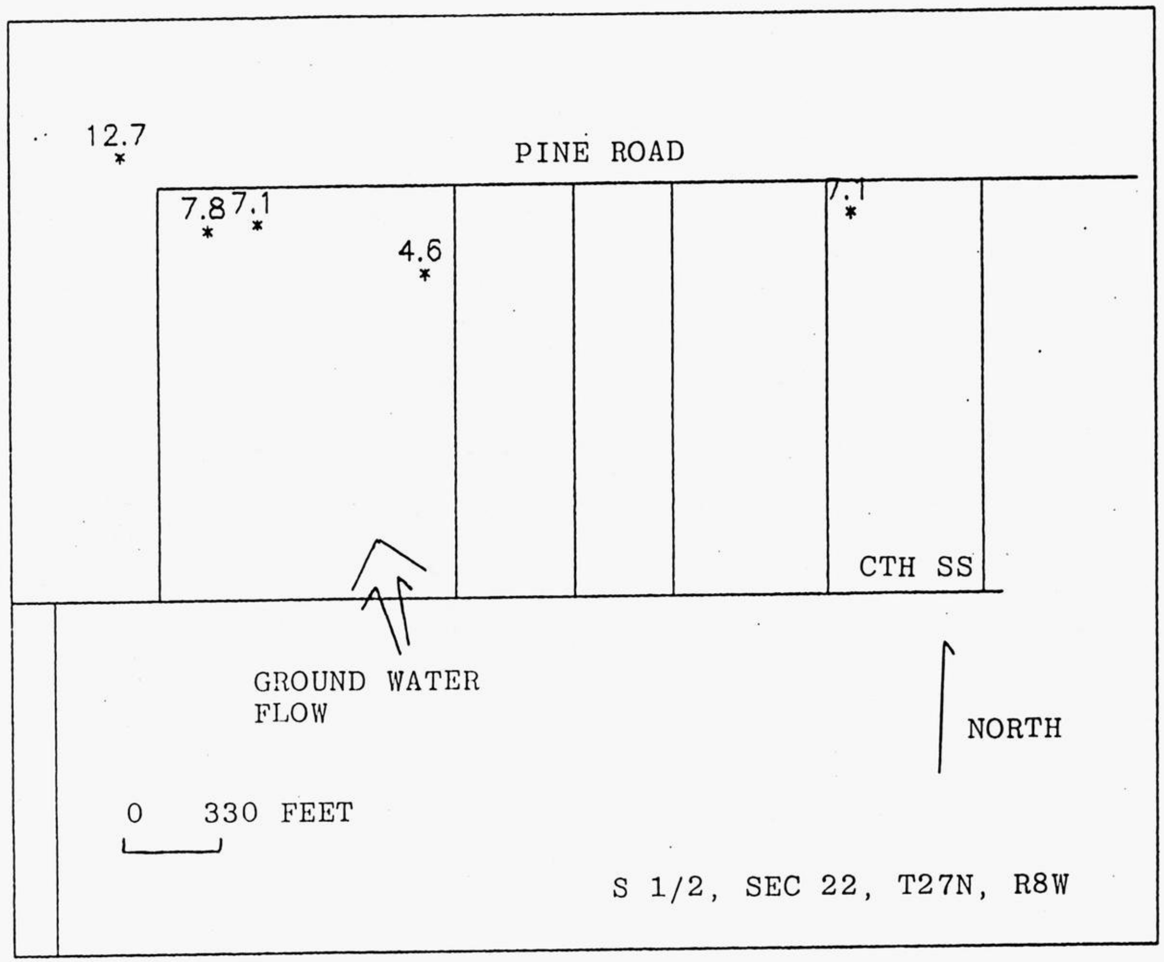

Figure 7. Nitrate-nitrogen in mg/L for private water-supply wells in Pine Grove-Deer Park subdivision, March 1990.

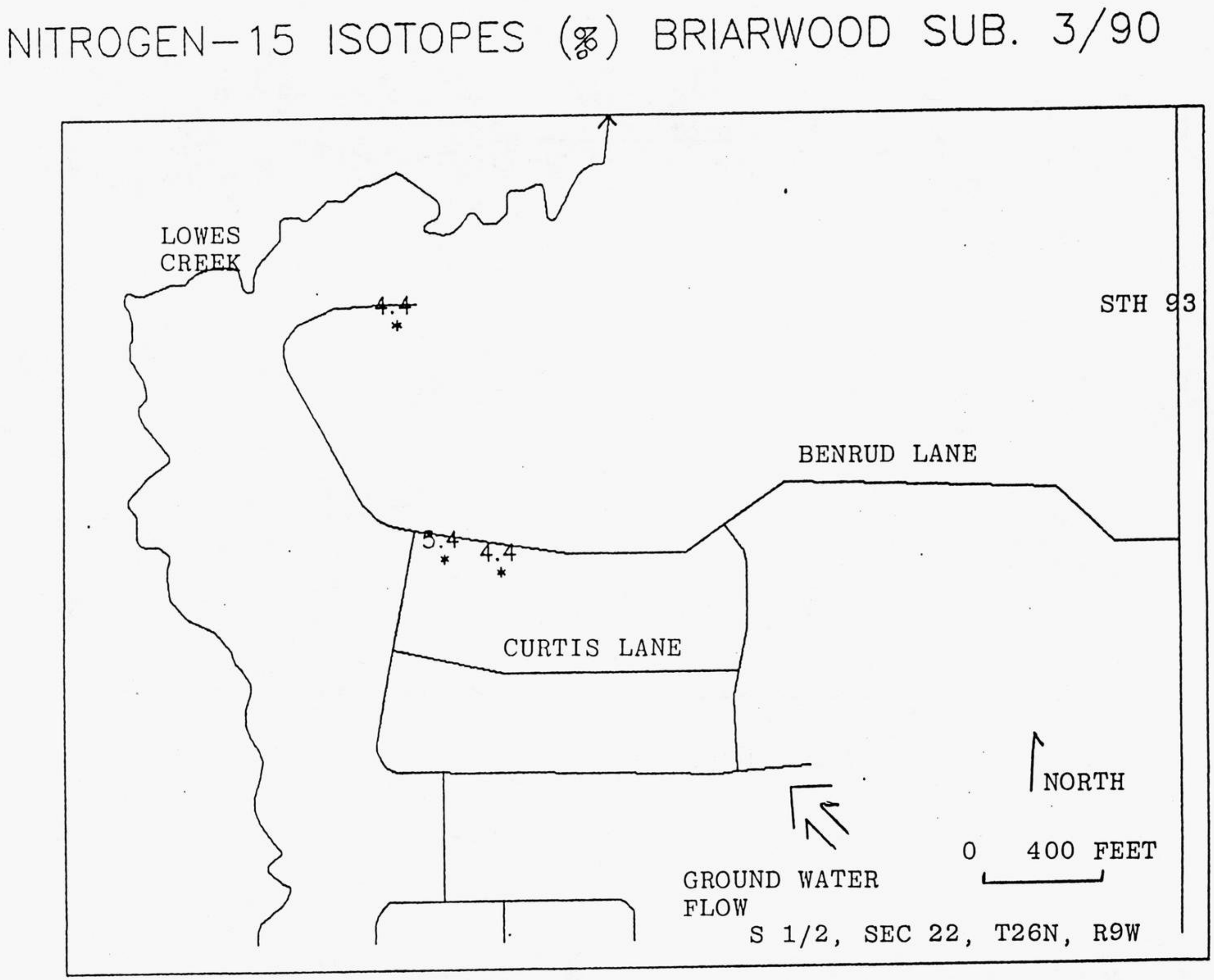

Figure 8. Nitrogen isotope values in parts per 10^3 differences, $\delta^{15}N$ for private water-supply wells in Briarwood subdivision, March 1990.

NITRATE—NITROGEN BRIARWOOD SUBDIVISION 3/90

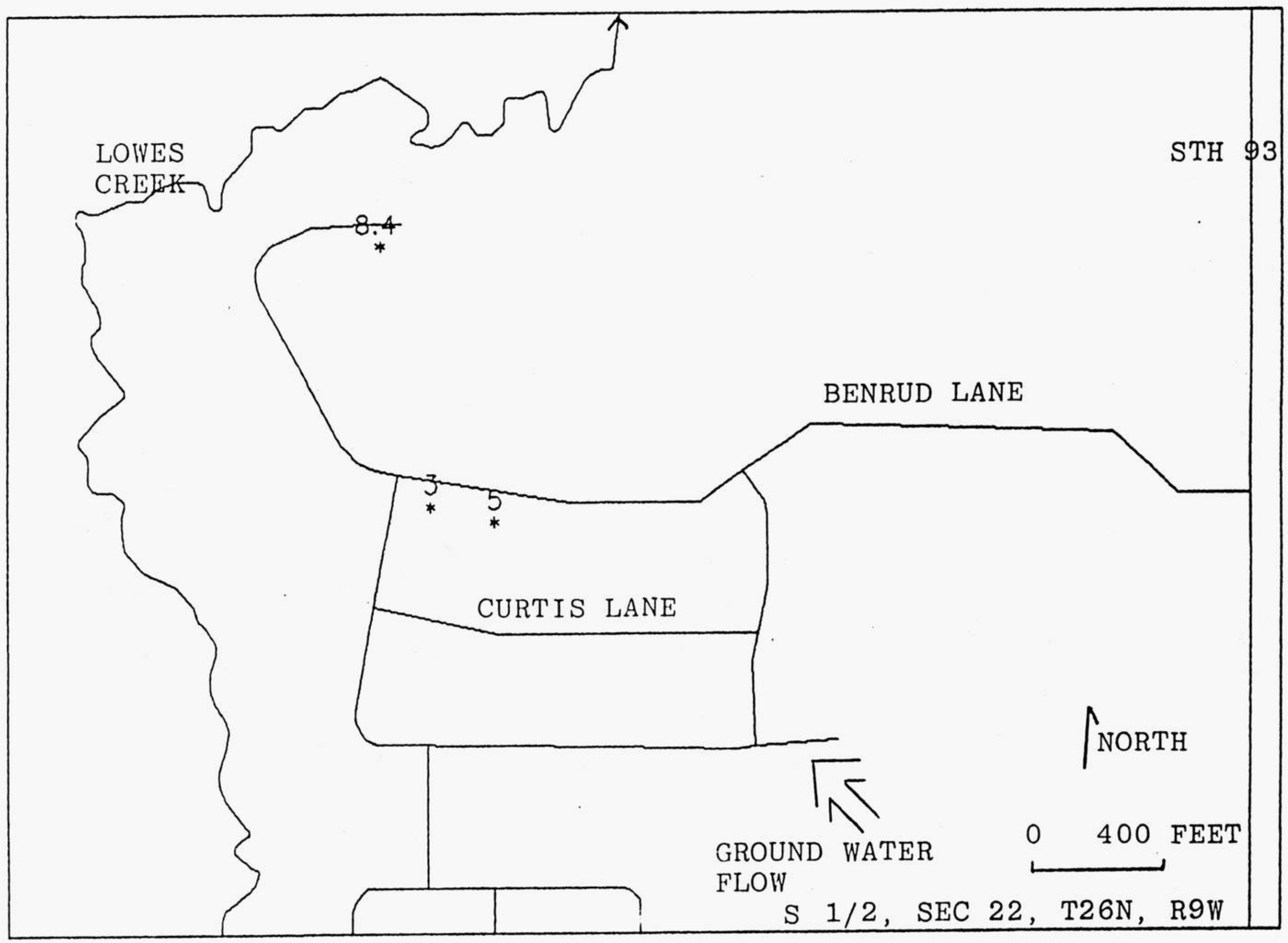

Figure 9. Nitrate-nitrogen in mg/L for private water-supply wells in Briarwood subdivision, March 1990.

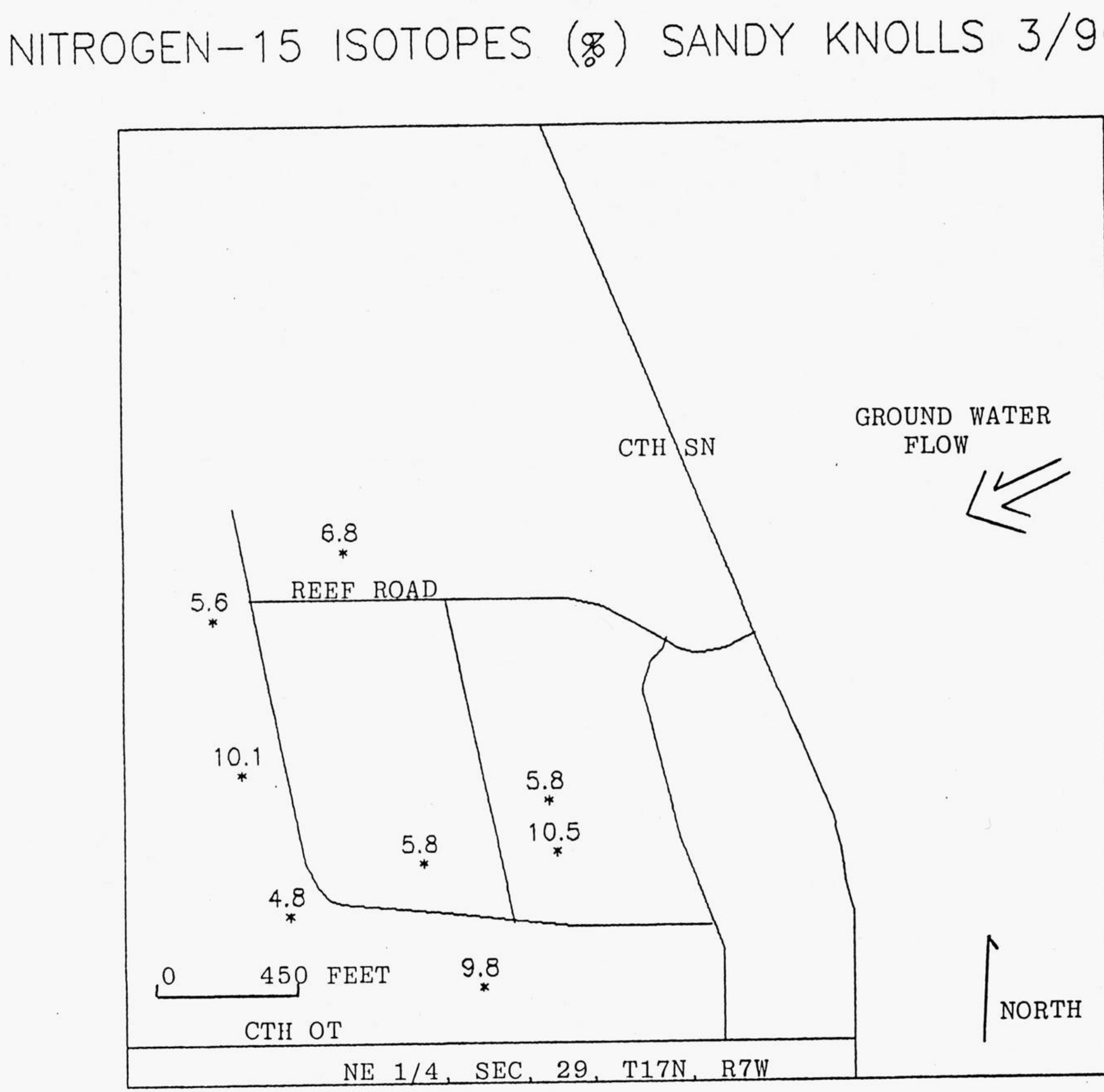

Figure 10. Nitrogen isotope values in parts per 10^3 differences, $\delta^{15}N$ for private water-supply wells in Sandy Knolls subdivision, March 1990.

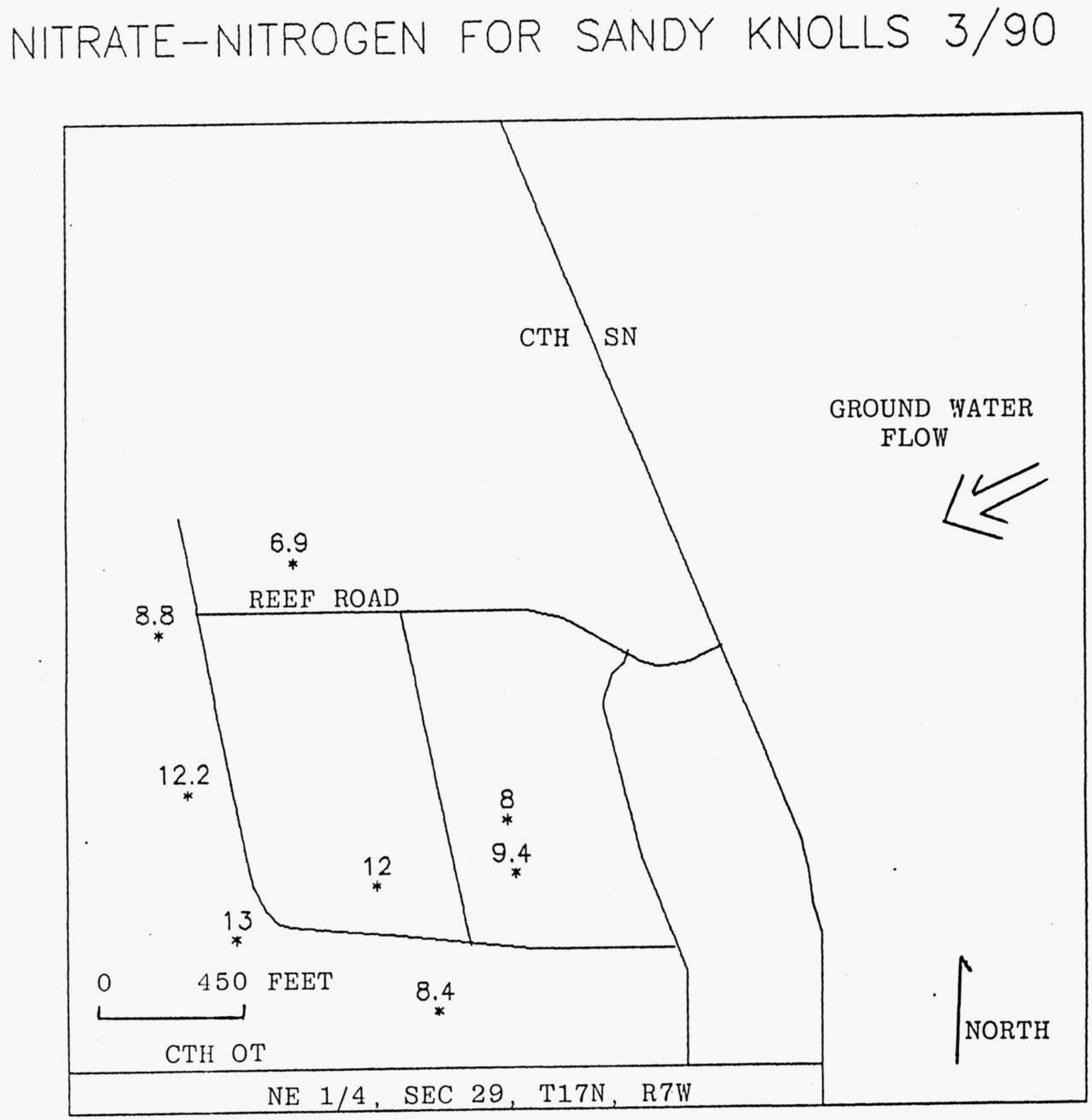

Figure 11. Nitrate-nitrogen in mg/L for private water-supply wells in Sandy Knolls subdivision, March 1990.

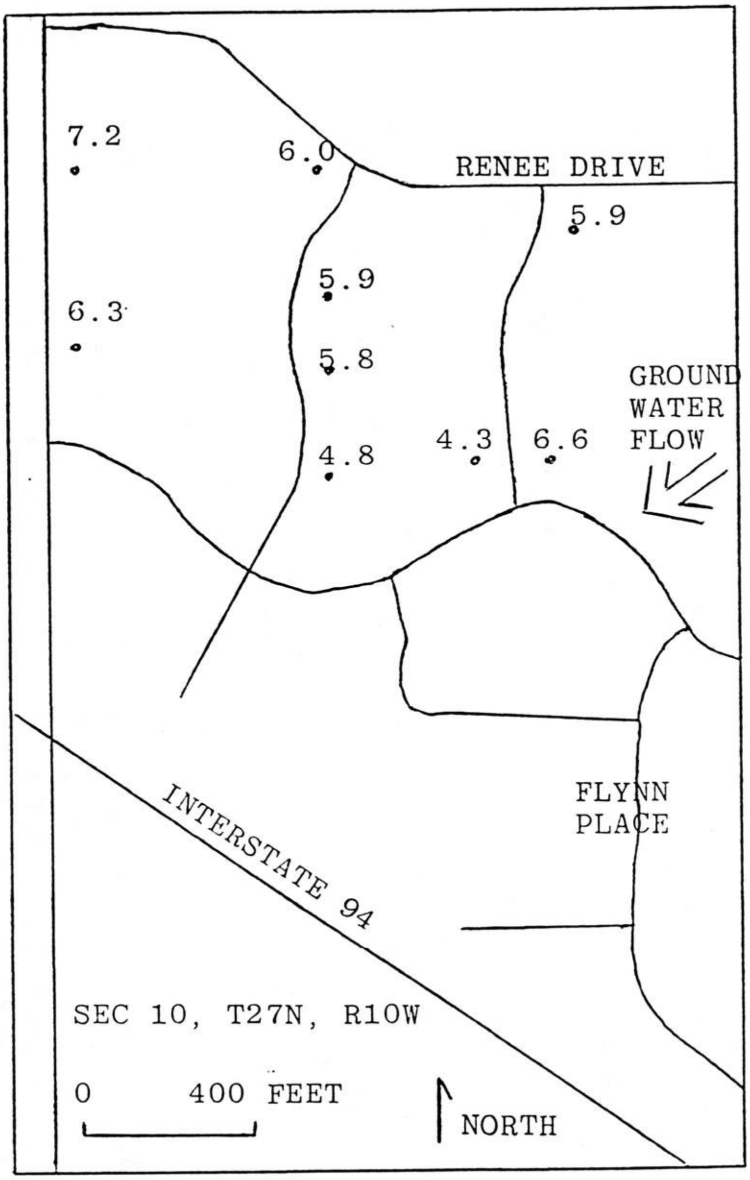

Figure 12. Nitrogen isotope values in parts per 10^3 differences, $\delta^{15}N$ for private water-supply wells in Mill Run subdivision, March 1990.

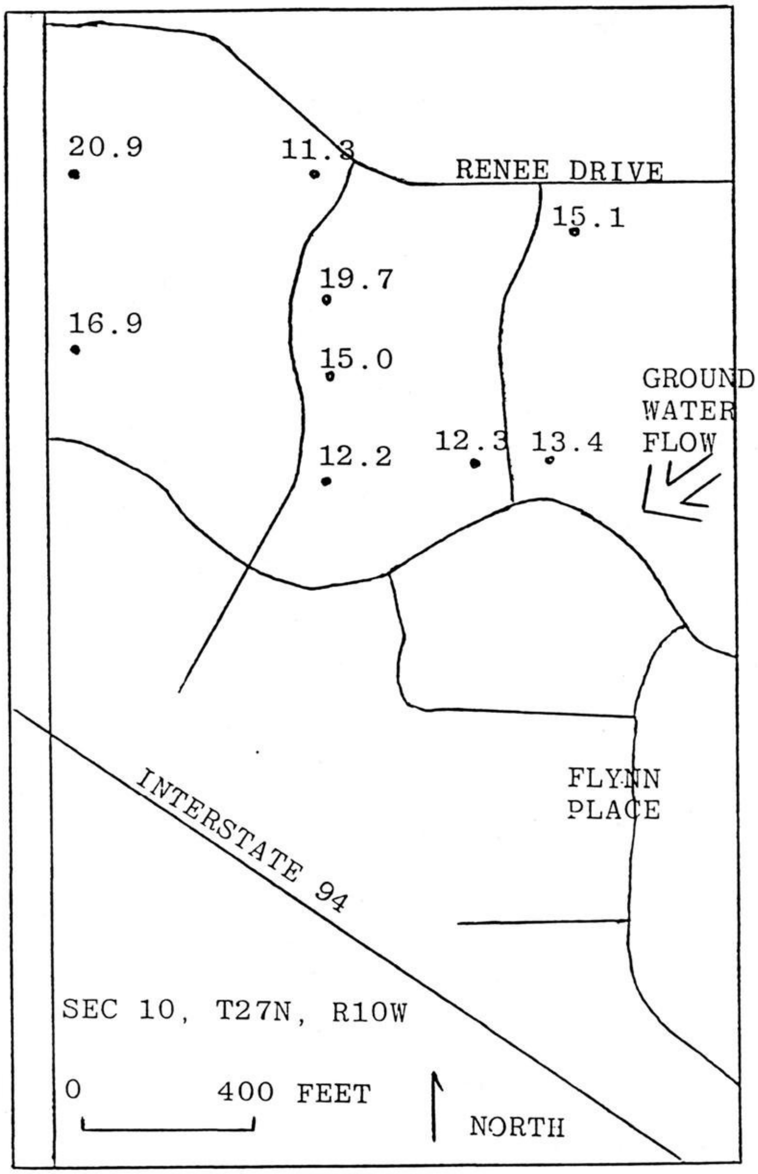

Figure 13. Nitrate-nitrogen in mg/L for private water-supply wells in Mill Run subdivision, March 1990.

TABLE 4. $\delta^{15}N$ ISOTOPE AND NO_3-N VALUES FOR EACH SUBDIVISION.

Subdivision	Sample No.	Parts per 10^3 difference, $\delta^{15}N$	mg/L NO_3-N
Sandy Knolls	1	9.8	8.4
	2	10.1	12.2
	7	6.8	6.9
	23	5.6	8.8
	24	10.5	9.4
	27	5.8	8.0
	39	5.8	12.0
	40	4.8	13.0
	mean	7.4 ± 2.3	9.8 ± 2.3
Oak Park	17	7.9	11.3
	18	6.8	13.0
	20	5.6	16.9
	33	8.5	6.0
	44	8.3	9.5
	45	5.2	12.0
	46	5.9	7.3
	47	5.8	12.0
	48	7.3	11.9
	mean	6.8 ± 1.2	11.1 ± 3.2
Mill Run	3	6.3	16.9
	4	5.9	19.7
	10	6.0	11.3
	11	4.3	12.3
	13	5.9	15.1
	14	6.6	13.4
	15	4.8	12.2
	19	7.2	20.9
	21	5.8	15.0
	mean	5.9 ± 0.8	15.2 ± 3.4
Pine Grove	5	6.3	7.1
	8	5.2	4.6
	12	5.2	7.8
	16	5.0	7.1
	22	5.5	12.7
	mean	5.4 ± 0.5	7.9 ± 3.0
Lowes Creek	29	6.0	4.6
	30	3.6	4.4
	mean	4.8 ± 1.7	4.5 ± 0.1
Briarwood	35	5.4	3.0
	36	4.4	8.4
	37	4.4	5.0
	mean	4.7 ± 0.6	5.5 ± 2.7

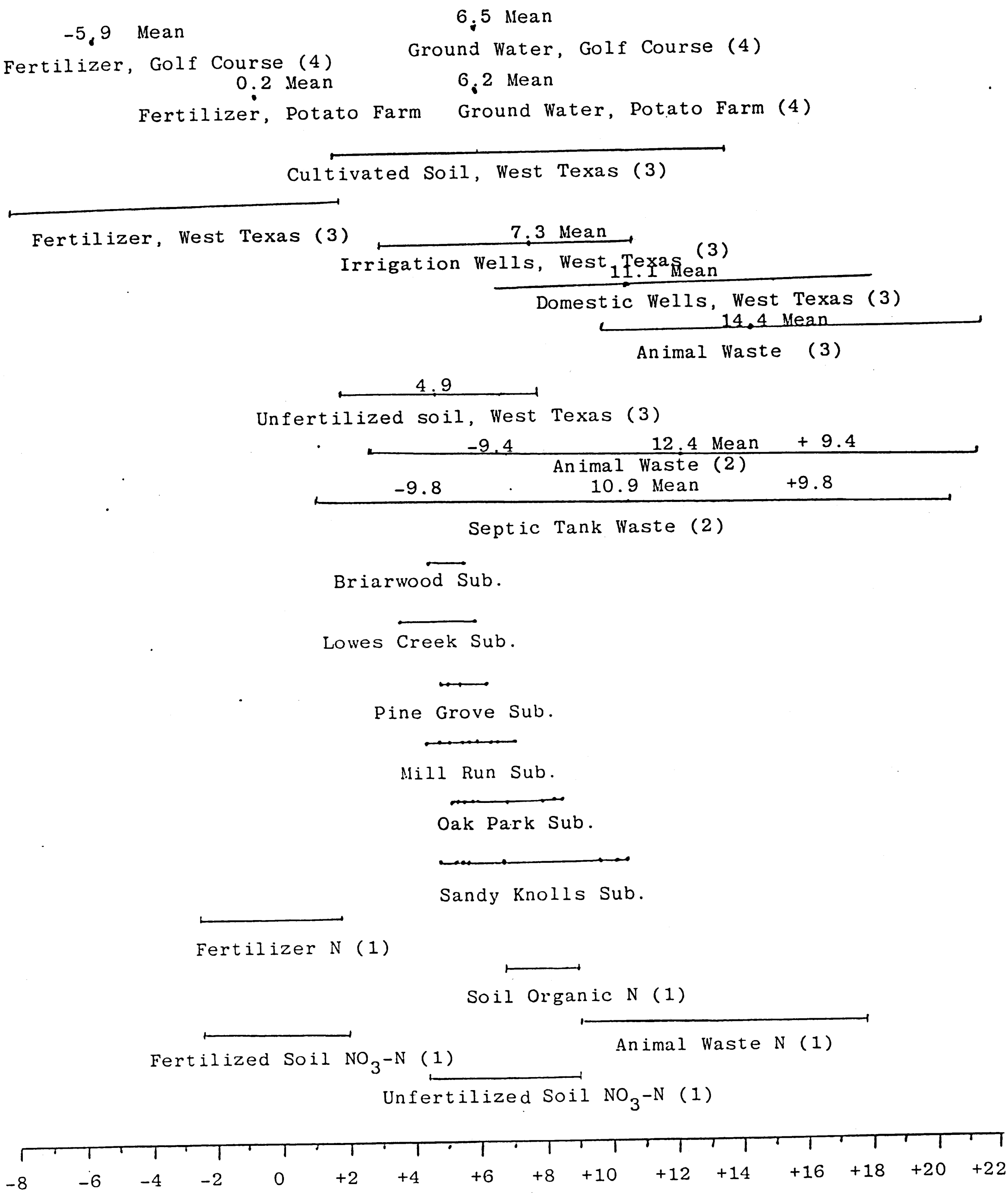

Figure 14. $\delta^{15}N$ ranges of potential nitrate sources and for Sandy Knolls, Oak Park, Mill Run, Pine Grove-Deer Park, Lowes Creek, and Briarwood subdivisions. References are (1) Gormly and Spalding (1979), (2) Wolterink et al. (1979), (3) Kreitler (1979), and (4) Flipse and Bonner (1985).

The following observations may be made from the data of Figure 14.

1. Fertilizer has a range of $\delta^{15}N$ lower than animal waste with no overlap of the range of values as presented by Gormly and Spalding (1979) and Kreitler (1979). However, Wolerink et al. (1979) present a wider range of $\delta^{15}N$ for septic tank waste and animal waste with the lower $\delta^{15}N$ values for septic tank waste slightly overlapping with the higher $\delta^{15}N$ values for fertilizer.

2. Unfertilized soil has a range of $\delta^{15}N$ isotope values generally between the range of values for fertilizer and animal waste.

3. Ground water beneath the potato farm and golf course in the Long Island study has higher $\delta^{15}N$ (6.2 and 6.5 respectively) than the fertilizer used at the earth's surface (0.2 and -5.9 respectively). Flipse, Jr. and Bonner (1985) attribute this increase in $\delta^{15}N$ to fractionation processes that occur within the soil during infiltration. Flipse, Jr. and Bonner (1985) also state that nitrogen-isotope ratios of fertilizer-derived nitrate were not altered to an extent that would make them indistinguishable from animal-waste-derived nitrates in ground water. However, this statement may assume that the lowest $\delta^{15}N$ for animal waste is greater than 10.

4. Cultivated land in West Texas has a wide range of $\delta^{15}N$ isotope values with the range wider than and not overlapping with the range for fertilizer used in West Texas. The range of $\delta^{15}N$ for cultivated land does overlap with the range of values for animal waste of West Texas.

5. The range of $\delta^{15}N$ (3.6 to 10.5) for the subdivisions in Wisconsin overlaps with the range of values for cultivated and unfertilized soil in Texas (Kreitler, 1975), unfertilized soil in Nebraska (Gormly and Spalding, 1979), septic tank waste of Wolterink et al. (1979) and, in part, with animal waste of Gormly and Spalding (1979).

Denitrification and volatilization are possible fractionation processes that increase $\delta^{15}N$ (Flipse, Jr. and Bonner, 1985). The denitrification of nitrate is subject to a large kinetic isotope effect (Delwiche and Steyn, 1970), which results in an increase in $\delta^{15}N$ in the remaining nitrate. The extent of denitrification in the vadose zone beneath the subdivisions is unknown. However, the sandy well drained soils which underlie the subdivisions are more likely to promote nitrification and less likely to promote denitrification than more clay rich poorly drained soils.

Volatile loss of gaseous ammonia is another possible cause of isotopic fractionation of fertilizer nitrogen applied in reduced forms (NH_4 and organic N). This process was found to be responsible for most of the ^{15}N isotope enrichment in nitrate produced from the nitrification of animal waste (Kreitler, 1975), and to be active when ammonium-based fertilizers were applied to basic soils (Kreitler, 1979). The extent of volatilization in the soil beneath or upgradient of the subdivisions is unknown.

The following are possible interpretations of the $\delta^{15}N$ data of Table 4 and Figure 14.

1. The $\delta^{15}N$ for the subdivisions indicate a source from fertilizers with isotope fractionation processes within the soil raising the $\delta^{15}N$ from less than 2 up to within the range of 3.6 to 10.5.

2. The $\delta^{15}N$ for the subdivisions indicate a septic tank waste source especially for the higher values (9.8, 10.1, and 10.5) in Sandy Knolls subdivision.

3. The $\delta^{15}N$ for the subdivisions indicate neither a fertilizer or septic-tank waste source but an unfertilized soil source.

4. The $\delta^{15}N$ for the subdivisions indicate some combination of a fertilizer source, septic tank waste source, or unfertilized soil source.

Conclusion

The $\delta^{15}N$ data for the water-supply well samples within the six subdivisions is inconclusive as to the source of nitrate-nitrogen in the wells.

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