# CHEMICAL, ISOTOPIC, AND MICROBIOLOGICAL EVIDENCE FOR DENITRIFICATION DURING TRANSPORT OF DOMESTIC WASTEWATER THROUGH A THICK UNSATURATED ZONE IN THE MOJAVE DESERT, SAN BERNARDINO COUNTY, CALIFORNIA

*By* Roy A. Schroeder, Peter Martin, *and* J.K. Böhlke

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### Conversion Factors

| Multiply | By | To obtain |
|---|---|---|
| acre | 0.4047 | hectare |
| foot (ft) | 0.3048 | meter |
| foot per day (ft/d) | 0.3048 | meter per day |
| gallon per day (gal/d) | 0.3785 | liter per day |
| mile (mi) | 1.609 | kilometer |

Temperature is given in degrees Celsius (°C), which can be converted to degrees Fahrenheit (°F) by the following equation:

$$°F=1.8(°C)+32.$$

### Abbreviations

mg/L: milligram per liter
$\mu M$: micromole per liter
permil: part per thousand
V-SMOW: Vienna Standard Mean Ocean Water

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## Abstract

Septic-tank wastewater disposed in 30-foot-deep seepage pits (dry wells) at 46,000 residences is estimated to equal about 18 percent of natural recharge to the sole-source aquifer beneath the upper Mojave River Basin, which is rapidly becoming urbanized, in the high desert northeast of Los Angeles. Nitrogen in the downward-infiltrating wastewaters represents a significant potential source of nitrate contamination to underlying ground water, but increases in nitrate concentration in ground water have not yet been observed. The low nitrate concentration in the ground water may be the result of lateral dispersion in the unsaturated zone, dilution below the water table, or denitrification of wastewater nitrate in the unsaturated zone.

Measured vertical rates of movement of wastewater wetting fronts through the unsaturated zone at three newly occupied residences ranged from 0.07 to 1 foot per day. Those measurements, along with moisture-content profiles at older residences, indicate that some wastewater has reached the water table beneath communities that are older than 5 to 10 years. As wastewater percolates from seepage pits into the unsaturated zone, reduced nitrogen is converted rapidly to nitrate at shallow depths. Analyses of water extracts of soil cores and of soil moisture from suction lysimeters deep beneath seepage pits at several residences indicate that nitrate concentrations commonly decrease with depth. The largest nitrate decreases seem to coincide with increased content of fine-grained sediments or proximity to the water table. Nitrate-reducing bacteria were found in soil cores collected from two residences. Between lysimeters at 160 and 199 feet at one residence, the decrease in nitrate concentration coincided with a large increase in sulfate, decrease in alkalinity, and increase in $\delta^{15}N$ in nitrate. Those data are consistent with denitrification by oxidation of iron sulfide to produce ferric oxides; but if such a reaction occurs, it must be in domains that are small in comparison with the sampled volumes because the waters also contain substantial quantities of dissolved oxygen. The predominantly low nitrate concentrations in the area's ground water are consistent with the operation of a nitrogen-removal mechanism, possibly denitrification, as wastewater moves through an unsaturated zone that averages 150 feet in thickness. However, the reducing capacity of the sediments to maintain denitrification is not known.


## INTRODUCTION

Septic-tank wastewater disposed in 30-foot-deep seepage pits (dry wells) at 46,000 residences in the communities of Victorville, Apple Valley, and Hesperia, is estimated to equal about 18 percent of natural recharge to the sole-source aquifer in the upper Mojave River Basin, which is rapidly becoming urbanized, in the high desert 80 mi northeast of Los Angeles (fig. 1).

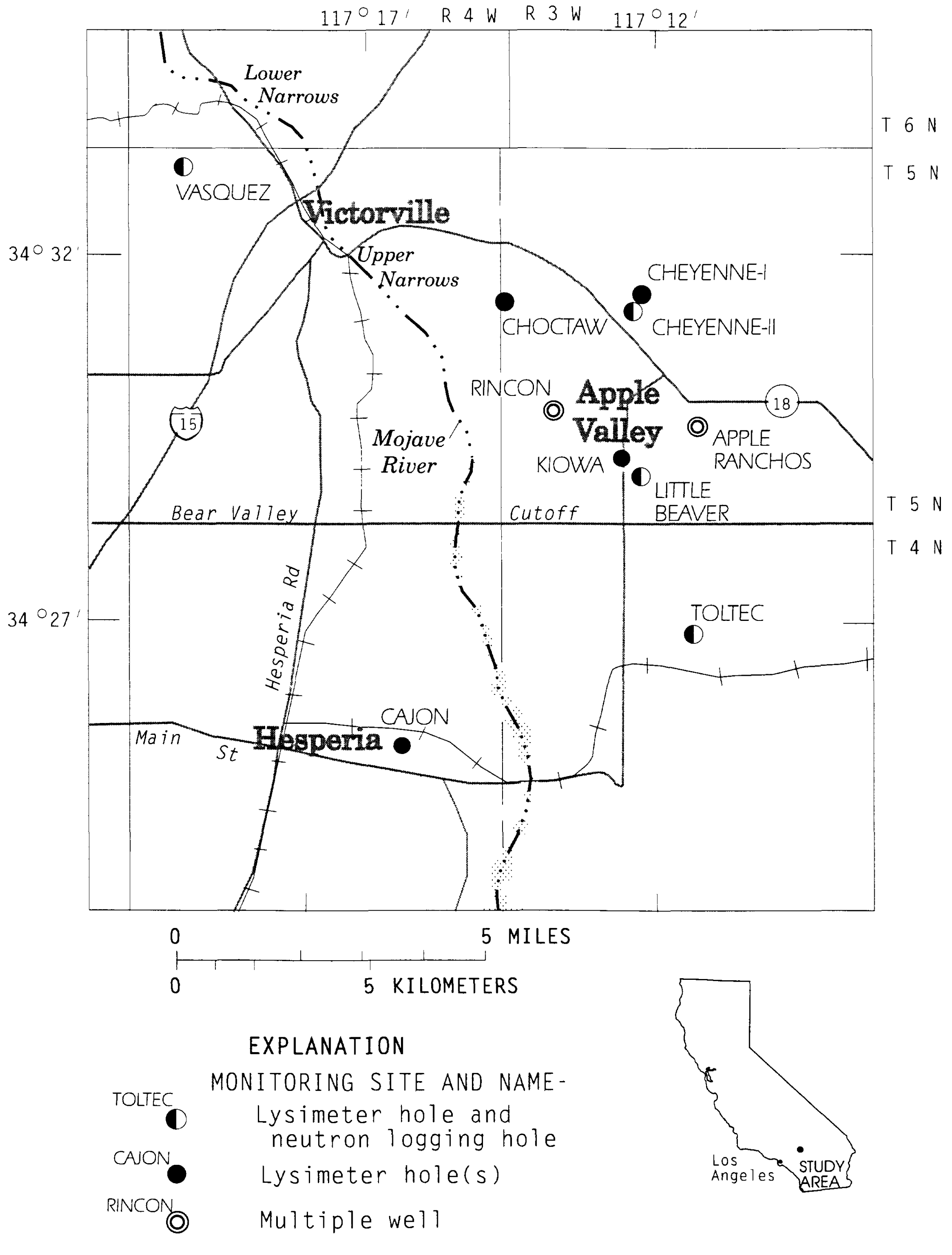


**Figure 1.** Location of study area and monitoring sites in the upper Mojave River Basin.

The use of seepage pits in this area, instead of near-surface leach fields that are more commonly used elsewhere, is necessitated by the widespread existence of an impermeable caliche layer near the land surface. This wastewater-disposal practice has caused concern about possible contamination of the underlying aquifer, especially by nitrate. In response to that concern, the U.S. Geological Survey began a study in 1988 to

1. measure vertical rates of wastewater movement through the unsaturated zone;
2. determine which chemical constituents of the wastewater reach the water table; and
3. evaluate the current and potential effects of wastewater discharge on future ground-water quality.

This report presents a summary of results of that study, which was funded jointly by the California Regional Water Quality Control Board, Lahontan Region, and the U.S. Geological Survey (Southern California Regional Aquifer-System Analysis program and other funds).

## INSTRUMENTATION AT EIGHT RESIDENCES

Monitoring was done to follow the movement of wastewater and to detect chemical changes in the water in the unsaturated zone at eight residences (locations shown in fig. 1) chosen to represent a range in geohydrologic conditions, residential density, and years of seepage-pit operation. Test holes were drilled next to the seepage pits at five previously occupied residences and through the bottoms of the seepage pits at three residences that were occupied soon after completion of drilling and instrument installation in 1988 (table 1). Cores were collected for analysis of various physical, chemical, and microbiological properties. Multiple-depth suction lysimeters were installed for water-quality sampling at all eight residences. Galvanized-steel access tubes were installed to measure changes in soil moisture by neutron logging at the three newly occupied residences and at one previously occupied residence (Cheyenne-I site).

**Table 1.** Characteristics of monitoring sites at eight residences in the upper Mojave River Basin

[gal/d; gallon per day; mg/L, milligram per liter; ft, foot; --, not determined]

| Site of residence | Year seepage pit constructed | Winter water usage, 1989 (gal/d) | Wastewater nitrogen (mg/L) | Depth to water table, 1988–89 (ft) |
|---|---|---|---|---|
| Toltec | 1988 | 388 | -- | [1]220 |
| Cheyenne-I | 1983 | 750 | 53 | 112 |
| Vasquez | 1988 | 250 | 26 | 85 |
| Choctaw | 1978 | 625 | 46 | 124 |
| Little Beaver | 1988 | 875 | -- | [1]150 |
| Kiowa | 1979 | 812 | 22 | [1]150 |
| Cheyenne-II | 1987 | 338 | 60 | 117 |
| Cajon | 1977 | 260 | 40 | 252 |

[1]Water table not reached by drilling; depth based on estimate from Subsurface Surveys, Inc., (1990, pl. 4).

## POSSIBLE SOURCES OF NITRATE

Downward-infiltrating domestic wastewater is considered to be the dominant potential source of nitrate contamination to ground water in the study area because

1. there are few domestic wells at which faulty well construction could allow wastewater to "short circuit" a thick unsaturated zone that ranges from less than 50 ft beneath the Mojave River to more than 400 ft in parts of Apple Valley and Hesperia;
2. landscaping, and the accompanying use of nitrogen fertilizers, is minimal in this desert environment;
3. what little agriculture exists has always been confined to a narrow strip adjacent to the Mojave River; and
4. although natural nitrate-rich soils are present in parts of the Mojave and Death Valley deserts, these soils generally are small in areal extent and highly localized, and none have been reported near the study area.

## OBSERVED NITRATE CONCENTRATIONS IN GROUND WATER

Nitrogen concentrations in wastewater samples from several residences averaged almost 50 mg/L (milligrams per liter), or 3,600 $\mu M$ (micromoles per liter), and the nitrogen in the wastewater was present mostly as ammonium and organic nitrogen (table 1). Nitrogen concentrations in infiltrating wastewater sampled below seepage pits were similar (typically 20 to 60 mg/L), but the nitrogen in the infiltrating wastewater was present mostly as nitrate, indicating that nitrification of wastewater occurs rapidly during infiltration. Despite this large potential source of downward-moving nitrate contamination, the nitrogen concentrations (mostly as nitrate) averaged only 1.2 mg/L (90 $\mu M$) in recent samples from 20 municipal wells in the study area. Ground water sampled just below the water table at three residences and between 5 and 100 ft below the water table in multiple-depth wells at the Apple Ranchos and Rincon sites (locations shown in fig. 1) also contains little nitrate.

Possible reasons for the absence of widespread nitrate contamination in the area's ground water include the following:

1. Wastewater has reached the water table but is diluted by large-scale vertical mixing in a saturated zone that is as much as 1,000 ft thick.
2. Wastewater has been retained within the unsaturated zone and has not yet reached the water table.
3. Nitrogen in the wastewater has been attenuated by processes such as denitrification as the wastewater moves through the unsaturated zone.

Each of these explanations was evaluated in this study.

# "PREDICTED" NITRATE CONCENTRATIONS IN GROUND WATER

Differences in major-ion characteristics at the Rincon multiple-depth wells indicate that vertical mixing with wastewater probably will occur within the upper 50 ft of the saturated zone. Results of simulations made using a single-cell mixing model similar to that described by Bauman and Schafer (1985) that allows nitrate from wastewater to be diluted by instantaneous vertical mixing indicate that readily measurable increases in nitrate concentration would be expected in shallow ground water within 10 years after the wastewater reaches the water table (fig. 2). The model simulates conditions that would be expected in the area between the Apple Ranchos and Rincon multiple-depth wells—a part of Apple Valley in which most of the houses were constructed in the early 1960's and where depth to ground water is sufficiently small that wastewater should have reached the water table long ago. The absence of high nitrate concentrations in the multiple-depth wells suggests that either nitrogen is being removed from the septic-tank wastewater or the vertical mixing depth must be substantially greater than 50 ft.

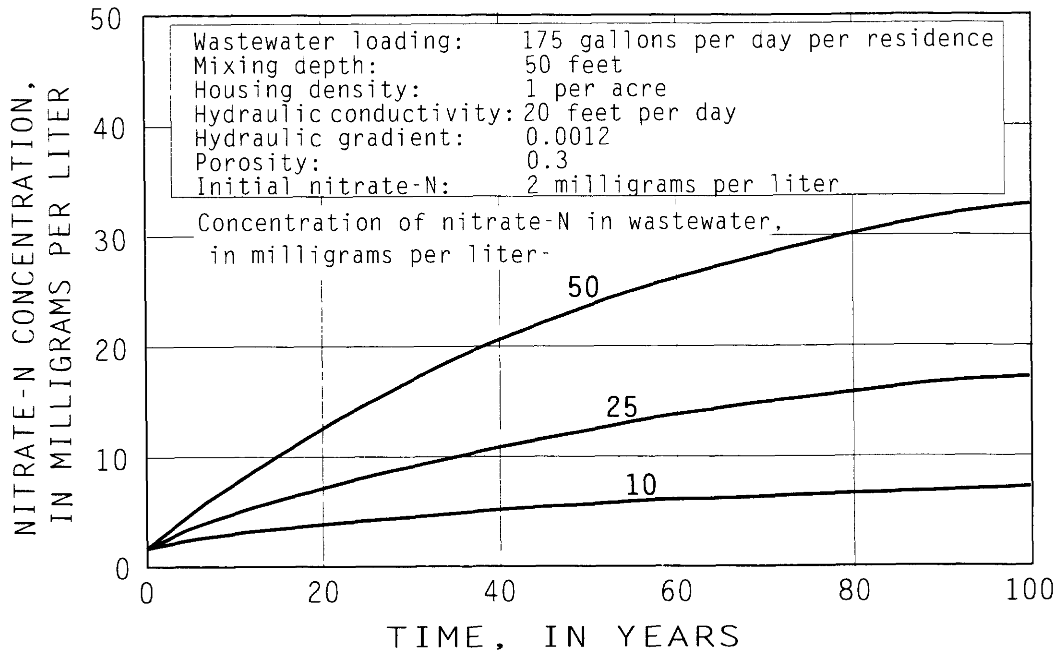


**Figure 2.** Model-computed nitrate concentrations in ground water in the study area for selected nitrate-N concentrations in wastewater.

## RATE OF WASTEWATER MOVEMENT

The arrival of the wastewater wetting front in the unsaturated zone was recognized from (1) temporal changes in neutron-log profiles; (2) the recovery of large sample volumes from lysimeters; and (3) increased soil moisture in cores. Downward movement of the front is indicated by the differences between successive neutron logs for a recently occupied residence at Toltec (fig. 3). Lysimeters at each depth at that site yielded recoverable water only after the neutron log indicated that the wastewater wetting front had arrived. Soil-moisture contents are relatively high throughout the unsaturated zone at the established sites and indicate that the wastewater wetting front has reached the water table at most older sites.

Measured rates (from neutron-log profiles and the suction lysimeters) of vertical movement of the wastewater wetting front in the upper 100 ft ranged from 0.07 to 1.0 ft/d at the three newly occupied residences. These measured vertical rates correspond to traveltimes of several months to about 6 years for wastewater to reach the water table at an average depth of about 150 ft (table 1). However, the vertical rates decrease with depth, possibly owing to lateral movement of the wastewater induced by stratigraphic barriers or increasing consolidation (or cementation) of the soils. Evidence for lateral movement was found in test holes drilled 25 ft from the seepage pit at the Cheyenne-I site and 50 ft from the seepage pit at the Choctaw site.

Further evidence for lateral movement can be seen from calculations of water volume potentially held within the unsaturated zone and from chemical data at the Cajon site. If the wastewater input is 260 gal/d (table 1), porosity is 0.3, and depth to ground water is 250 ft, then wastewater would saturate the soil in a 5-foot cylinder directly underlying the seepage pit in less than 2 months—if one assumes no lateral movement. However, chemical data (table 2, mainly chloride concentrations) indicate that wastewater at the Cajon site has moved to a depth of approximately 200 ft in about 15 years. If uniform lateral movement is assumed, then wastewater would saturate the soil within a cylinder to a radial distance of about 30 ft after 15 years. Actual moisture content of soils in the wastewater zone is somewhat less than saturation, and some pre-existing moisture is present in the soils; therefore, wastewater movement would be proportionately faster and lateral movement a little farther (it varies inversely with the square root of

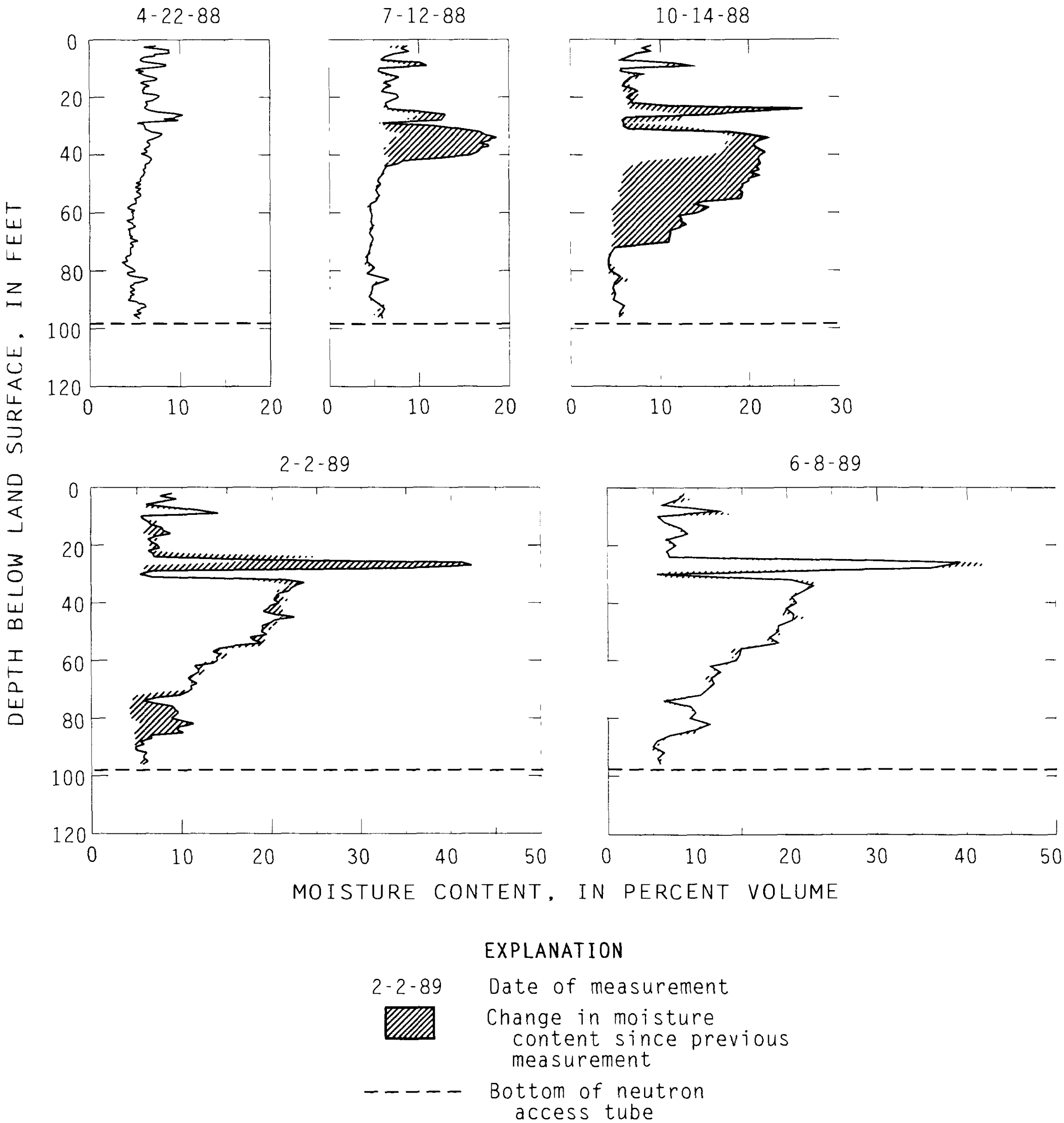


**Figure 3.** Temporal variation in soil-moisture content in the unsaturated zone at the Toltec site. Shaded areas indicate moisture additions in succeeding time intervals and record the downward movement of the wastewater wetting front.

moisture content) than indicated by the calculations, but still much less than the average distance between seepage pits at adjacent residences.

Although wastewater has not yet reached the water table at the Cajon site, abundant soil moisture is present beneath the wastewater zone as evidenced by large sample volumes from the lysimeter at 243 ft. This might be the result of an accumulation of natural soil moisture in advance of the wastewater wetting front in a manner similar to that described by Young and others (1992). Specific-conductance measurements at the Cajon site between November 1991 and March 1993 indicate that no systematic

**Table 2.** Water-quality data from lysimeters at the Cajon residence, Hesperia, California

[Specific conductance, in microsiemens per centimeter at 25° Celsius, is given as a range of values from as many as 11 samples collected November 1991–March 1993 at each depth. Isotope ratios are in permil deviation from standards (V-SMOW for O and H, air for N, Pee Dee Belemnite for C). All other constituent concentrations except pH are in μ*M* (micromole per liter), and most are from single samples. If more than one sample was analyzed, the median value is given. --, no data; <, actual value is less than value shown]

| Depth below land surface, in feet | 35 | 70 | 97.5 | 130 | 160 | 199 | 243 | 257 |
|---|---|---|---|---|---|---|---|---|
| **Constituent or property** | | | | | | | | |
| Specific conductance | 448-553 | 435-525 | 463-544 | 470-590 | 493-567 | 391-535 | 248-315 | 1,000-1,330 |
| Delta deuterium | -- | -- | –59.5 | -58.0 | -61.0 | -59.0 | -59.2 | -54.5 |
| Delta inorganic carbon-13 | -12.75 | -9.50 | -11.35 | -- | -12.40 | -- | -10.30 | -- |
| Delta nitrate nitrogen-15 | 10.5 | 9.5 | 8.2 | 7.8 | 7.8 | 14.8 | -- | -- |
| Delta oxygen-18 | -- | -- | -8.75 | -8.65 | -8.85 | -8.85 | -8.90 | -7.50 |
| Molecular oxygen | 120 | 140 | 140 | -- | 190 | 160 | 220 | -- |
| pH (standard units) | 7.2 | 7.4 | 7.1 | 7.4 | 7.4 | 4.0 | 7.2 | 3.5 |
| Hydrogen ion | .06 | .04 | .08 | .04 | .04 | 100 | .06 | 320 |
| Calcium | 820 | 720 | 770 | -- | 320 | 340 | 250 | -- |
| Magnesium | 260 | 300 | 360 | -- | 490 | 370 | 280 | -- |
| Sodium | 2,700 | 2,100 | 2,200 | -- | 1,600 | 1,800 | 1,400 | -- |
| Potassium | 280 | 40 | 30 | -- | 50 | 90 | 50 | -- |
| Alkalinity (as $CaCO_3$)[1] | 700 | 550 | 350 | 560 | 450 | –34 | 660 | –140 |
| Sulfate | 110 | 120 | 120 | -- | 150 | 620 | 540 | -- |
| Chloride | 1,600 | 1,200 | 1,300 | -- | 1,000 | 890 | 110 | -- |
| Fluoride | 20 | 10 | 10 | -- | 10 | 10 | 10 | -- |
| Bromide | .6 | .5 | .8 | -- | .8 | .6 | .6 | -- |
| Silica | 1,300 | 1,300 | 800 | -- | 730 | 1,500 | 890 | -- |
| Nitrite | 2 | <1 | <1 | 1 | <1 | <1 | 1 | <1 |
| Ammonia | 7 | 11 | 4 | 24 | 31 | 64 | 3 | 80 |
| Nitrite plus nitrate | 1,900 | 1,900 | 2,100 | 2,400 | 2,500 | 1,600 | 21 | 10 |
| Organic nitrogen | 40 | 20 | <10 | 40 | 20 | 10 | <10 | 60 |
| Orthophosphate | 110 | 74 | 100 | 2 | 5 | <1 | <1 | 1 |
| Boron | 18 | 10 | 7 | -- | <1 | 2 | 3 | -- |
| Chromium | -- | -- | -- | -- | -- | <.02 | <.02 | -- |
| Copper | -- | -- | -- | -- | .1 | 5 | .06 | -- |
| Iron | .16 | <.05 | <.05 | -- | <.05 | 9 | 18 | 8,400 |
| Lead | -- | -- | -- | -- | .02 | .14 | .01 | -- |
| Manganese | .02 | .13 | <.02 | -- | .04 | 12 | 11 | 1,800 |
| Nickel | -- | -- | -- | -- | .02 | .5 | .02 | -- |
| Zinc | -- | -- | -- | -- | 14 | 7 | .7 | -- |
| Organic carbon | 240 | 300 | 230 | 350 | 290 | 170 | 450 | 1,200 |

[1]Negative alkalinity calculated relative to a titration endpoint of pH = 4.5.

change occurred in the chemical compositions in either the five lysimeters within the wastewater zone (above 199 ft) or the lysimeters at 243 and 257 ft that are below the wastewater zone. During the same period, the specific conductance at 199 ft increased monotonically in 11 successive samples by a total of nearly 40 percent, presumably because the wastewater was displacing some residual low-conductance and low-chloride soil moisture. The lysimeter at 257 ft has yielded very little water since 1991 when the water table dropped below this depth in response to nearby pumping and the recent drought in California.

## EVIDENCE FOR NITRATE REDUCTION IN THE UNSATURATED ZONE

The possibility that nitrate is being removed from wastewater in the unsaturated zone is indicated by decreases in nitrate concentrations with depth in deionized-water core extracts and in lysimeter samples from the Cheyenne-I site (fig. 4) and in lysimeter samples from the Cajon site (table 2). Additional evidence for nitrate reduction comes from the observation that bacteria capable of nitrate reduction are present in some of the soils. Higher numbers of nitrate-reducing bacteria and reduced nitrate concentrations seem to be associated with finer grained sediments and higher moisture contents at the Cheyenne-I site (fig. 4). The nitrate-concentration profile from lysimeter samples at the Cheyenne-I site is based on monitoring for a period of only about 1 year in 1988-89, after which the lysimeters were covered by a concrete driveway. The concrete was removed in the spring of 1993 and monitoring was resumed to ascertain if the decrease in nitrate concentration at depth that is shown in figure 4 represents transient or more permanent conditions, and to further evaluate possible reasons for the concentration profile that was observed.

If the reduced nitrate concentration at 199 ft at the Cajon site is solely the result of mixing of high-nitrate wastewater (represented by data at 160 ft) with low-nitrate residual soil moisture (represented by data at 243 ft), then the chloride data indicate that the lysimeter at 199 ft contains 88 percent wastewater. That percentage predicts that nitrate concentration should be 2,200 $\mu M$. However, data in table 2 show that the actual nitrate concentration is only 1,600 $\mu M$.

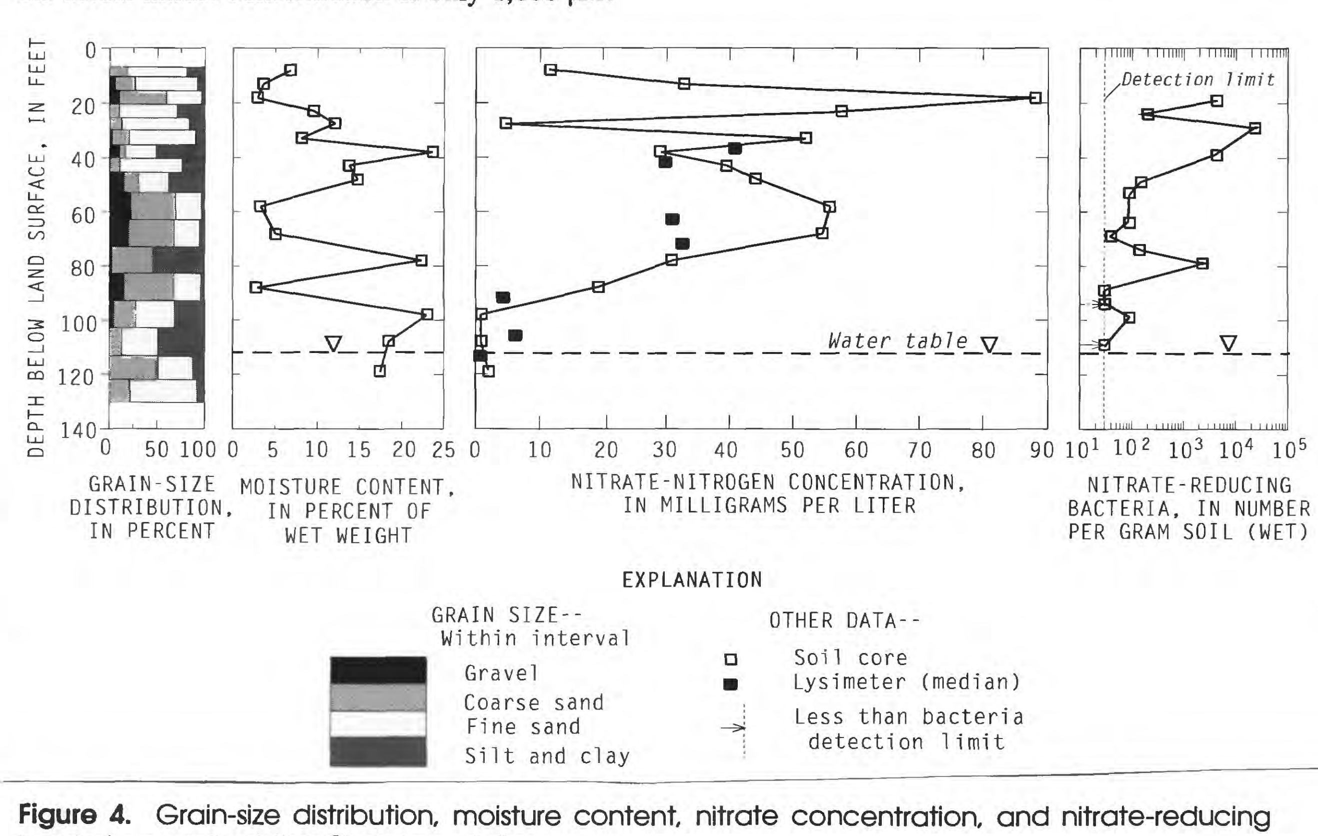


**Figure 4.** Grain-size distribution, moisture content, nitrate concentration, and nitrate-reducing bacteria counts at the Cheyenne-I site.

Isotopic evidence for denitrification is provided by the nitrogen-15 enrichment of the nitrate in a sample from the lysimeter at 199 ft in comparison with samples from the shallower lysimeters (fig. 4; table 2). The relation between the isotope ratios and nitrate concentrations is consistent with kinetic isotope fractionation during nitrate-consuming reactions with an isotope-enrichment factor of about -10 permil. That enrichment factor is within the wide range of values (approximately -30 to -5 permil) previously associated with denitrification in a variety of ground-water environments (Hübner, 1986; Mariotti and others, 1988; Böttcher and others, 1990; Schroeder and others, 1991; Smith and others, 1991). Isotope-enrichment factors at the lower end of this reported range, in this and in other studies, could indicate the influence of specific physicochemical factors on the reaction, or mixtures of unreacted and reacted waters from differing physicochemical environments (Hübner, 1986; Mariotti and others, 1988).

If it is assumed that waters entering the lysimeters at 160 and 199 ft both are dominated by wastewater that had the same initial composition (both having high chloride concentrations, for example), then the differences between their measured compositions can be treated as coefficients in a net chemical reaction that occurs between those depths as the wastewater moves downward. If that assumption is correct, then the data in table 2 indicate that the wastewater gained approximately 470 $\mu M$ of sulfate and 970 $\mu M$ of hydrogen ion (including alkalinity lost to carbonic acid) while losing 900 $\mu M$ of nitrate. If those quantities are normalized to hold chloride constant (assuming minor dilution of wastewater by low-salinity soil water, represented by data from the lysimeter at 243 ft), then the corresponding gains and losses are approximately 550 $\mu M$ sulfate and 980 $\mu M$ hydrogen ion for 700 $\mu M$ nitrate. Those coefficients and underlying assumptions have large uncertainties; nevertheless, they can be interpreted as evidence for a reaction involving denitrification by oxidation of iron sulfide (table 3, reaction 1). Iron sulfide minerals have not been analyzed in the unsaturated-zone sediments, but they could be present in small quantities.

The data are not supportive of major reactions involving oxidation of organic carbon (reaction 4) or ferrous silicates or oxide minerals (reaction 3). The estimated ratios of sulfate gain to nitrate loss between 160 and 199 ft at the Cajon site (0.52 and 0.79, for raw data and chloride-normalized data, respectively) are similar to the ratio required by reaction 1 (0.67). The estimated ratios of hydrogen-ion gain to nitrate loss (1.1 and 1.4) are larger than that required by reaction 1 (0.33) and could indicate additional hydrogen-ion contributions from reactions such as 5 to 8 (table 3), if mixtures of waters from diverse soil domains were sampled.

**Table 3.** Stoichiometries of some possible redox and precipitation reactions at the Cajon residence, Hesperia, California

| Geochemical reaction | Electron donors | Electron acceptors |
|---|---|---|
| 1. $6NO_3^- + 2FeS_2 + 2H_2O \rightarrow 3N_2 + 2FeOOH + 4SO_4^{2-} + 2H^+$ | Fe,S | N |
| 2. $14NO_3^- + 5FeS_2 + 4H^+ \rightarrow 7N_2 + 10SO_4^{2-} + 5Fe^{2+} + 2H_2O$ | S | N |
| 3. $2NO_3^- + 10FeO + 4H_2O + 2H^+ \rightarrow N_2 + 10FeOOH$ | Fe | N |
| 4. $4NO_3^- + 5(CH_2O) + 4H^+ \rightarrow 2N_2 + 5H_2CO_3 + 2H_2O$ | C | N |
| 5. $4FeS_2 + 15O_2 + 10H_2O \rightarrow 4FeOOH + 8SO_4^{2-} + 16H^+$ | Fe,S | O |
| 6. $2FeS_2 + 7O_2 + 2H_2O \rightarrow 4SO_4^{2-} + 2Fe^{2+} + 4H^+$ | S | O |
| 7. $4Fe^{2+} + O_2 + 6H_2O \rightarrow 4FeOOH + 8H^+$ | Fe | O |
| 8. $Fe^{3+} + 2H_2O \rightarrow FeOOH + 3H^+$ | — | — |

Denitrification generally is inhibited in the presence of oxygen, whose concentration in soil-moisture samples brought to the surface by nitrogen-gas pressurization of the lysimeters ranged from 120 to 220 $\mu M$ (3.7 to 7.1 mg/L). If denitrification occurs at the Cajon site, the presence of oxygen in the lysimeter samples indicates that it could be occurring in domains (possibly microenvironments) that are smaller than the volumes sampled by the lysimeters.

Denitrification by iron sulfide oxidation has been reported in ground waters from saturated-zone environments (Postma and others, 1991; Korom, 1992). If such a reaction occurs at the Cajon site, it is relatively deep in the unsaturated zone near the wastewater front. It is not known whether this is a stable feature controlled by the occurrence of some unique lithology or mineralogy localized in sediments at that depth, or if it is a transient feature that will move with the wastewater front and eventually allow nitrate to reach the underlying ground water. Monitoring is continuing at the Cajon residence to identify initial arrival of wastewater in the lysimeters at 243 and 257 ft and to determine if the apparent nitrate-sulfate exchange reaction observed near 199 ft is a stable phenomenon associated with the aquifer or a more transient condition associated with the advancing wastewater front.

## SUMMARY

Physical and chemical evidence indicates that wastewater from domestic disposal systems has reached the water table beneath many of the older residences in the upper Mojave River Basin. However, any increases in ground-water nitrate concentration that might have resulted from the wastewater disposal have not yet been detected. The absence of high observed nitrate concentrations in the area's ground water may be partly explained by the failure of recent wastewater discharge to have reached the water table, by dilution through vertical mixing within the saturated zone, and by the scarcity of monitor wells perforated solely near the water table. There also is some chemical and isotopic evidence locally for denitrification of wastewater nitrate within the unsaturated zone; however, the distribution of denitrification, and the capacities of the soils for maintaining it, are not known. Effects of wastewater on the aquifer could be detected by future monitoring in both the saturated and unsaturated zones.

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