Tracing Sources of Nitrate in the Long Island Aquifer System

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Abstract

Several public supply wells in the Town of Huntington, located along the north shore of Long Island, New York, have been closed because the nitrate concentration exceeds the public drinking water standard. Although the area was once extensively farmed, most of it was converted to subdivisions in the early 1950’s as urbanization spread eastward from New York City. To determine what action should be taken to reduce nitrate contamination of the aquifer, it is important to assess to what extent the problem is due to present land use versus past agricultural activities. Samples were collected from shallow monitoring wells screened near the water table to characterize the geochemistry of groundwater associated with various types of land use. The geochemistry of the public-supply wells was compared to that of the monitoring wells and geochemical data from the literature. The nitrogen and oxygen isotopes of groundwater nitrate were used in conjunction with other geochemical data to place constraints on potential nitrate sources. The \( \delta^{18}O \) values of the public supply wells indicate that the nitrate is primarily derived from nitrification of ammonium in the soil. The \( \delta^{15}N \) values suggest that cultivation sources predominate; however, the influence of septic system wastes can be seen in the elevated \( \delta^{15}N \) values of some of the shallower well fields in the area. Since application of manure to fields might also produce elevated \( \delta^{15}N \) values, additional geochemical criteria were used to distinguish water contaminated by residential as opposed to agricultural land use. Although conducted on Long Island, this study is expected to have broader significance since urbanization of agricultural lands can be expected to place similar pressures on the development of water resources worldwide.

Introduction

The widespread contamination of groundwater with nitrate is of global concern to both environmentalists and public health officials. The Long Island aquifer system presents a unique challenge to water resource managers because there are 3 million people in Nassau and Suffolk Counties who live on top of this unconfined sole-source aquifer. Nitrate contamination of groundwater on Long Island stems from a number of different land use activities [Perlumutter and Koch, 1972; Kreitler et al., 1978; Katz et al., 1980; Porter, 1980; Flipse et al., 1984; Flipse and Bonner, 1985]. Potential sources of nitrate include: a) agricultural fertilizers; b) turf grass fertilizers; c) septic tank effluent or leaking sewer lines; d) landfill leachate; e) commercial or industrial wastewater; and f) acid rain. The objective of this study is to place constraints on the relative contributions of various land use activities to nitrate contamination of the aquifer.

Most of Long Island was once intensively farmed, however, the importance of agriculture waned as urbanization spread eastward from New York City. To evaluate actions that might be taken to reduce nitrate contamination in Long Island
groundwater, it is crucial to be able to distinguish between contributions of nitrate due to previous agricultural activities versus those due to urbanization and present land use practices.

Previous research has demonstrated that $\delta^{15}N_{\text{nitrate}}$ and isotopic composition of groundwater nitrates can be used to help distinguish among different nitrate sources [Aravena et al., 1993; Durka et al., 1994; Wassenaar, 1995; Ging et al., 1996; Kendall et al., 1997]. In addition, the oxygen isotopes can be used to identify processes, such as denitrification, that may alter the concentration and isotopic composition of nitrate [Amberger and Schmidt, 1987; Böttcher et al., 1990]. Isotopic data can be used more effectively when combined with additional geochemical data [Böhlke and Denver, 1995]. Supplemental geochemical data provides additional constraints necessary to distinguish between nitrates that are likely to be derived from residential as opposed to agricultural land use.

To assess the impact of different types of land use, groundwater samples were collected from both monitoring wells and public supply wells on Long Island. Monitoring wells are generally 6 inches or less in diameter and have short screens near the water table. Samples from these wells were used to characterize the chemistry of the groundwater and the isotopic composition of nitrates associated with particular land uses. Public supply wells are generally 10 to 16 inches in diameter and are screened over 20-80 foot intervals at various depths within the aquifer. Samples from these wells were used to determine the average concentration and isotopic composition of nitrates present within a cross-section of the aquifer. The geochemistry of the public supply wells was compared to that of the monitoring wells and other information obtained from the literature to place constraints on the relative importance of various sources of nitrate contamination in the aquifer.

**Description of Study Area**

The Northport study area lies within the Town of Huntington, which is located along the north shore of Long Island (Figure 1). Several of the shallower public supply wells in this area have already been closed because the concentrations of nitrate exceed the public drinking water standard of 10 mg/l of nitrate-N. At least one sample was collected from each of the public supply well fields currently in operation in the Northport area. Monitoring well samples were collected primarily from residential areas within the Town of Huntington. In addition, one residential sample was collected from a private well in the north shore community of Rocky Point and an agricultural sample was obtained from eastern Long Island. The data are summarized in Table 1. Land use in Suffolk County has been quantified and mapped by the Long Island Regional Planning Board [1982]. Figure 2 shows land use in the vicinity of the public supply wells.

Residential groundwater is expected to contain a mix of nitrates derived from lawn fertilizers and septic system wastes. Low to medium density residential development (1-4 dwellings/acre) accounts for almost 90% of the residential land use in the Town of Huntington. A small network of sewers exists in downtown Northport; however, most wastewater is disposed of through domestic cesspools or septic systems. According to a household fertilizer use survey [Koppelman, 1978], the average nitrogen fertilizer application rate in the Town of Huntington is 66 kg/acre/year. This is almost 50% greater than the average rate for Long Island as a whole [Koppelman et al., 1984].
Huntington also has the third highest per capita residential water consumption of the towns in Suffolk County (SCDHS, 1987).

Older water in some of the deeper wells may contain nitrates derived from previous agricultural activities. The only major agricultural operation left in the Northport area today is Richter’s Orchards; however, aerial photos from 1947 [Suffolk County Department of Health Services] show that much of the study area was once under cultivation. By 1956, most agricultural land had been converted to subdivisions [Huntington Town Planning Board, 1956].

Field and Analytical Procedures

Temperature, pH, specific conductance, and dissolved oxygen concentrations were measured in the field. Dissolved oxygen concentration was also determined by Winkler titration on return to the lab at Stony Brook. The alkalinity of the monitoring well samples was determined by field titration. The Suffolk County Water Authority lab determined the alkalinity of the public supply well samples. For these samples, total alkalinity was assumed to be conservative and carbonate alkalinity was adjusted for changes in pH that occurred between the field and the lab.

All samples were field filtered through 0.45 micron flow-through filters and stored on ice until they could be refrigerated or frozen on return to the lab. Carbon isotope samples were collected in glass septum bottles. Samples for the determination of $\delta^{15}N_{nitrate}$ and $\delta^{18}O_{nitrate}$ were preserved with mercuric chloride.

Sulfate, nitrate and chloride concentrations were determined by ion chromatography at Stony Brook. Nitrate for isotopic analysis was extracted on ion-exchange columns after precipitation of sulfate with barium chloride. $\delta^{15}N_{nitrate}$ values and $\delta^{18}O_{nitrate}$ values were determined by the Environmental Isotope Lab (EIL) at the University of Waterloo, Ontario [Aravena et al., 1993]. Carbon isotope analyses were also performed by EIL. Anonymous replicates were submitted to the lab. Replicates were within ±0.2 ‰ for nitrogen in nitrate, ± 0.1 ‰ for carbon in DIC, and ± 1.0 ‰ for oxygen in nitrate.

Samples collected at the same time by Scott Meyerdierks of the Suffolk County Water Authority (public supply wells) and Ralph Milito of the Suffolk County Department of Health Services (monitoring wells) were submitted to their respective labs for analysis. Samples were analyzed for major and trace-element concentrations and a suite of pesticides and organic compounds.

Hydrogeology

The hydrogeology of Long Island is described in detail elsewhere [Jensen and Soren, 1971; Franke and McClymonds, 1972; Smolensky et al., 1989]. The Long Island aquifer system consists of a thick wedge of unconsolidated deposits, which rests on a crystalline bedrock surface that dips gently to the southeast. The Lloyd aquifer (sand), the Raritan confining unit (clay), and the Magothy aquifer (deltaic gravels, sand, silt, and clay) are Cretaceous Atlantic Coastal Plain deposits. The Upper Glacial aquifer consists of Pleistocene morainal deposits, outwash, and marine and lacustrine clays.
[Lubke, 1964]. The Upper Glacial aquifer rests unconformably on the irregular erosion surface of the Magothy aquifer except along the north shore where the Magothy has been completely eroded. The Magothy and the Upper Glacial aquifers are primary sources of public drinking water.

The Northport study area is located to the north of the groundwater divide. Buxton and Modica [1992] have modeled groundwater flow along a north/south transect near the Nassau/Suffolk border approximately 15 miles to the west (Figure 3). An approximation of the pattern and rate of groundwater flow in the Northport area can be obtained from this model. This pre-pumpage model predicts a maximum age of 250 years for water at the base of the Magothy north of the groundwater divide. Most of the groundwater in the study area is probably less than 100 years old.

North shore communities on Long Island are particularly vulnerable to nitrate contamination of their water supply because of aquifer characteristics north of the groundwater divide. In this area, much of the Magothy has been removed by erosion. The Upper Glacial aquifer contains thick sequences of highly permeable glacial outwash sediments that have a vertical hydraulic conductivity of 23 feet per day as compared to less than 1 foot per day for the Magothy aquifer [Buxton and Modica, 1992]. The high conductivity of Upper Glacial sediments facilitates the drawdown of contaminants into deeper parts of the aquifer. In addition, dissolved-oxygen measurements of groundwater in the Northport area indicate that the entire flowpath may be oxygenated. Aerobic conditions preclude the removal of nitrate by denitrification. Since nitrate behaves conservatively under these conditions, the isotopic composition of nitrate can be used to trace the source of the nitrate.

The residence time of water in the aquifer to the north of the groundwater divide is short compared to areas south of the divide due to the thinning of saturated sediments from south to north across Long Island [Franke and McClymonds, 1972]. The withdrawal of water by the pumping of public supply wells significantly reduces the residence time of water in this part of the aquifer system (Figure 4). As a result, groundwater quality can rapidly deteriorate as younger, more contaminated, water is drawn into deeper parts of the aquifer.

Nitrogen Loading According to Land Use

Nitrogen loading on Long Island has been studied in detail by the Long Island Regional Planning Board [Koppelman, 1978; Koppelman et al., 1984]. Table 2 summarizes nitrogen loading in the Town of Huntington according to land use. (See Bleifuss [1998] for a discussion of nitrogen loading studies on Long Island and the assumptions on which these data are based.) Both agricultural and residential land use are capable of producing concentrations of nitrate in groundwater that exceed the public drinking water standard. Concentrations of nitrate-N exceeding 10 mg/l are found in agricultural areas where farmers apply more than an average amount of fertilizer and in medium and higher density unsewered residential neighborhoods. The percentage of nitrogen derived from synthetic fertilizers as opposed to human or animal wastes varies as a function of land use. This variation will later be used to help place constraints on the potential sources of nitrate in the public supply wells.
Nitrogen and Oxygen Isotopes in Nitrate

The $\delta^{15}$N_{nitrate} and $\delta^{18}$O_{nitrate} values can be used to distinguish between different sources of nitrate. Figure 5 depicts fields for various nitrate sources based on the work of previous researchers. Due to the large oxygen isotopic contrast between nitrates produced in the atmosphere and those produced by microbial processes in the soil (nitrification), the oxygen isotopes in nitrate are particularly useful for the identification of nitrate from fertilizer [Amberger and Schmidt, 1987] and atmospheric nitrates [Durka, et al., 1994; Kendall et al., 1997]. The $\delta^{18}$O of nitrates produced by nitrification varies regionally because one oxygen in the nitrate is derived from oxygen gas and two oxygens are derived from soil water or groundwater [Anderson and Hooper, 1983; Hollocher, 1984].

On Long Island, $\delta^{18}$O for nitrates produced by nitrification of ammonium would be expected to range from approximately +2.5 to +3.2‰ assuming an isotopic composition of +23.5‰ for atmospheric oxygen [Amberger and Schmidt, 1987] and -7 to -8‰ for Long Island groundwater [U. S. Geological Survey Watstore data]. The $\delta^{18}$O_{nitrate} values in the public supply wells indicate that most nitrate is derived from nitrification of ammonium (Figure 6). Furthermore, they suggest that denitrification, which results in a 2:1 enrichment in the heavier isotope of the nitrogen and oxygen isotopes respectively [Amberger and Schmidt, 1987; Böttcher et al., 1990], is not an important process in this part of the aquifer. Measurements of dissolved oxygen support this conclusion. Denitrification takes place under anaerobic conditions or in environments in which oxygen availability is severely limited [Korom, 1992]. The public supply well samples range from 58 to 100% saturated with respect to dissolved oxygen.

Based on the oxygen isotope data, nitrate fertilizers do not appear to be an important nitrate source in the public supply wells in the Northport area. Ammonium nitrate, ammonium sulfate, and urea were the three nitrogen fertilizers heavily in use in neighboring Nassau County during 1920-1950 [Perlmutter and Koch, 1972] the same period of time during which land use in the Northport area was predominantly agricultural. If the nitrate from the ammonium nitrate fertilizer contributed significantly to the nitrate leached from agricultural land, then it would be possible to distinguish agricultural nitrates from other nitrate sources based on their enriched $\delta^{18}$O_{nitrate}. Since none of the public supply wells show substantially enriched $\delta^{18}$O_{nitrate} values, nitrate fertilizers can be dismissed as a significant source of nitrate in this area.

Variations in $\delta^{15}$N_{nitrate}

The $\delta^{15}$N_{nitrate} depends in part on that of the nitrogen source, however, this original signature may be modified by exchange with soil nitrogen [Komor and Anderson, 1993] and by ammonia volatilization [Kreitler, 1979; Flipse and Bonner, 1985] and denitrification [Mariotti, 1988; Böttcher et al., 1990]. Although ranges for various nitrogen sources can be obtained from the literature [Kreitler, 1975; Kreitler and Jones, 1975; Heaton, 1986], it is important to determine the isotopic composition of nitrates produced under local conditions.
The range of $\delta^{15}\text{N}_{\text{nitr}}$ observed by Kreitler and others [1978] in agricultural areas on Long Island (+2.7 to +8.8‰) overlaps with that reported by Flipse and others [1984] for seweraged residential neighborhoods (+1.1 to +7.7‰). Nitrates associated with cultivation practices can be derived from fertilizer or the breakdown of soil organic matter. In Long Island's coarse-textured aerobic soils, ammonia volatilization is probably the primary process driving the variation in the isotopic composition of soil nitrates. Flipse and Bonner (1985) attribute the relatively heavy $\delta^{15}\text{N}_{\text{nitr}}$ observed at golf courses (median +6.0‰) and potato fields (median +5.5‰) to ammonia volatilization prior to nitrification of ammonium and synthetic nitrogen fertilizers. Samples with depleted $\delta^{15}\text{N}_{\text{nitr}}$ are probably derived from the breakdown of organic matter. In this study, nitrates collected downgradient from the Smithtown waste transfer station, presumably derived from the breakdown of organic matter in the landfill, had a $\delta^{15}\text{N}_{\text{nitr}}$ of +3.2‰.

Groundwater in unsewered residential neighborhoods should contain a significant amount of nitrogen contributed by septic system wastes. Therefore, it should be possible to distinguish nitrates derived from agricultural land use from those derived from residential land use in the Northport Area. The disposal of wastewater through septic systems introduces isotopically heavy nitrogen to groundwater in residential areas. Aravena and others [1993] report a median $\delta^{15}\text{N}_{\text{nitr}}$ of +9.8‰ for nitrate downgradient from a domestic septic system in rural Ontario. In neighboring Nassau County, a median $\delta^{15}\text{N}_{\text{nitr}}$ of +7.8‰ was reported for Upper Glacial wells believed to be contaminated by wastewater [Kreitler et al., 1978].

The N-isotopic composition of nitrate in residential monitoring wells sampled during this study ranges from +6.2 to +8.4‰ with a median of +6.3‰. A variety of wells were sampled, some of which were downgradient from areas of intensive turf grass cultivation such as golf courses (A, E) and school playing fields (B) and others that were believed to be contaminated by septic system wastes (F, G, I). Nitrates in the wells influenced primarily by turf grass cultivation were isotopically lighter (+6.2, +6.2 +6.5‰) than those influenced by septic system wastes (+7.1, +7.1, +8.4‰). The lighter N-isotopic composition of the Huntington wells in comparison to those of Nassau County reflects the greater importance of turf grass fertilizers as a source of nitrate in the Town of Huntington. Since Huntington is less densely populated, one would expect to see a greater contribution of nitrate from turf grass fertilizers as opposed to septic system wastes.

It has been suggested that older agricultural waters on Long Island might also contain enriched $\delta^{15}\text{N}_{\text{nitr}}$ due to the application of manure as fertilizer [Kreitler et al., 1978; Porter, 1980]. Additional geochemical criteria will be introduced to help distinguish between agricultural and residential groundwater. It should be noted, however, that the application of manure to agricultural soils does not necessarily produce isotopically heavy nitrates. Aravena and others [1993] report a median isotopic composition of +4.6‰ for nitrates in agricultural soils fertilized with a combination of solid cattle manure and inorganic nitrogen fertilizers. The heavier $\delta^{15}\text{N}_{\text{nitr}}$ of the Magothy aquifer observed by Kreitler and others (1978) could also be explained by the drawdown of water from the Upper Glacial aquifer which has been contaminated by wastewater [Bleifuss, 1998].
Major Ion Chemistry of Residential and Agricultural Water

The major ion chemistry of groundwater varies according to land use. Ternary diagrams of the major cations and major anions show that agricultural and residential waters plot in different fields with little overlap (Figures 7a,b). Agricultural land use produces a calcium-sulfate type water due to liming and the application of fertilizers. Residential land use produces water that is more enriched in sodium and either chloride or bicarbonate. The high concentration of sodium and chloride in residential areas is due in part to an increased contribution of road salt due to the higher density of roads and in part to the elevated concentrations of these ions found in septic system plumes. High bicarbonate concentrations are also common within wastewater.

The monitoring-well samples collected during this study are plotted in Figures 7c,d. Except as indicated these samples were collected in residential neighborhoods. Most samples plot within the residential field with a few notable exceptions. Wells downgradient from golf courses or other areas of intensive turf grass cultivation, such as school playing fields, show an enrichment in calcium and magnesium which approaches that observed in agricultural areas. This is due to the application of lime to increase pH and dolomite lime to increase pH and supply magnesium. Intensive turf grass cultivation tends to obscure the differences between residential and agricultural waters. On the anion plot, an acidic waste contaminant plume with a high concentration of sulfate plots within the agricultural field. Sulfate is a common constituent of detergents and it is also used in cleaning cesspools.

The only public-supply wells that clearly show the enrichment in calcium and sulfate characteristic of agricultural land use are those at Gun Club Road (GC1, GC3). These wells plot just outside the agricultural field on both ternary diagrams (Figures 7e,f). The $\delta^{15}$N$_{\text{nitrate}}$ in these wells (+5.93‰, +6.03‰) is consistent with an agricultural source. In addition, these are the only wells in the study area that contain traces of pesticide (1-2 dichloropropane). Present land use in the vicinity of the wells is agricultural.

Sources of Nitrate in the Public Supply Wells

The lightest $\delta^{15}$N$_{\text{nitrate}}$ values are found in the two wells with the lowest concentration of nitrate (WA3, LH1). The Washington Street well is screened in the discharge zone and probably contains some of the oldest water in the study area. It is saturated with respect to dissolved oxygen, has a low specific conductance (32.7 $\mu$ S/cm), and contains less sulfate than modern rainwater, indicating that the water is close to "pristine" [Schoonen and Brown, 1994]. The light $\delta^{15}$N$_{\text{nitrate}}$ in this well (+4.3‰) suggests some contribution of nitrate from soil organic matter. Both the Washington Street and the Laurel Hill wells are located in low density residential neighborhoods, although there is some institutional and agricultural land use upgradient from the Laurel Hill site. The better than average water quality in these wells appears to be due to the older age of the water in the wells in combination with the lower density development in the immediate vicinity of the wells, which lessens the impact of the drawdown of younger water.

The $\delta^{15}$N$_{\text{nitrate}}$ values of public supply wells in the Northport area indicate that these
wells are dominated by nitrates derived from cultivation practices. The $\delta^{15}N_{\text{nitrate}}$ values of the public supply wells are consistent with low to medium density residential development, however, they could also be explained by mixing of older agricultural water with younger residential water. In deep wells that are screened close to the discharge zone (such as RS2) one would expect there to be some contribution from agricultural activities based on the approximate age of the water.

It seems unlikely that the nitrate contamination of public supply wells in the Northport area is primarily due to previous agricultural activities. Ternary plots of the major ions indicate that the well field at Gun Club Road is the only area where nitrate contamination can be clearly linked to intensive agricultural practices. Present land use in the vicinity is agricultural.

Although older agricultural waters might also contain nitrates with heavier isotopic compositions, additional geochemical data can distinguish between residential and agricultural waters. Elevated sodium concentrations are associated with residential land use while elevated calcium concentrations are associated with agricultural land use. Septic plumes introduce organic carbon into the aquifer which subsequently leads to a depletion in dissolved oxygen due to the respiration of organic carbon. In contrast, soil cultivation practices are intended to produce more aerobic conditions within the soil. Residential samples collected from monitoring wells show a positive correlation of $\delta^{15}N$ with sodium concentration (Figure 8a) and a negative correlation with dissolved oxygen concentration (Figure 8b). These same trends can be observed in public supply well samples (Figures 8c,d). Therefore, this heavier nitrogen is probably associated with septic system wastes rather than fertilization of agricultural fields with manure.

Several of the shallower wells in the study area clearly show the influence of septic system wastes. The public supply wells most enriched in $\delta^{15}N_{\text{nitrate}}$ are Waterside Road (WS2, +7.1‰) and Middleville Road (MR2, +6.8‰). At the Middleville Road site, chloride and sulfate concentrations have been reduced about 25% by reinjection of system water over the winter. The $\delta^{15}N_{\text{nitrate}}$ would likely have been higher before reinjection began since the median $\delta^{15}N_{\text{nitrate}}$ of wells in the area is +6‰ and the least contaminated wells (with lower isotopic compositions) are pumped in the winter. The shallower wells in both of these well fields have already been closed because the concentrations of nitrate exceed the drinking water standard. The positive correlation of $\delta^{15}N$ with sodium concentration and the negative correlation with dissolved oxygen suggests these nitrates are more likely to come from septic system wastes than application of manure to agricultural fields. The heavier carbon isotopic composition of DIC in these wells is also consistent with this interpretation. Medium to intermediate density residential neighborhoods are located upgradient from the Waterside well. Onsite sewage treatment at the Veteran's Administration Hospital may impact the Middleville Road well, especially since this is an old facility. These are among the shallowest well fields in the area and are the most likely to be influenced by recent land use activities.

The high concentration of nitrate found in public supply wells in the Northport area is surprising given the relatively low population density. In part, the problem can be attributed to aquifer characteristics, but turf grass cultivation practices probably exacerbate the situation. Due to its position north of the groundwater divide, the
residence time for water in this part of the aquifer is on the order of tens of years even without pumping. The high rate of withdrawal due to above-average water consumption accelerates the drawdown of contaminated water into deeper parts of the aquifer where the supply wells are screened. On average, 755 gallons of water per acre per day are discharged to groundwater by residents [SCDHS, 1987]. The natural recharge rate is 1720 gallons per acre per day [Peterson, 1987]. Consequently, up to 30% of the recharge in residential neighborhoods may consist of previously contaminated water. Aerobic conditions preclude the removal of nitrate through the process of denitrification.

Nitrogen loading calculations suggest that more than 50% of the nitrate in residential groundwater in the Northport area may be derived from turf grass cultivation. The $\delta^{15}$N$_{\text{nitrate}}$ values of residential monitoring wells supports this conclusion. The Suffolk County Department of Health Services [1987] found that the average nitrogen concentration in shallow monitoring wells in the Town of Huntington during the period 1973-1982 was 7.4 mg/l, the highest average observed for any town in Suffolk County during their investigation. Although previous agricultural activities may contribute nitrate to water in some of the deeper wells, the nitrate contamination problem cannot be expected to disappear as this older water discharges to Long Island Sound. Nitrates in some of the shallower well fields in the study area can be linked to residential land use. The closure of the shallower wells in these fields clearly demonstrates that changes in residential land use practices are necessary to protect the quality of groundwater in the Northport area.

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