A GEOCHEMICAL STUDY OF NITRATE CONTAMINATION IN THE LONG ISLAND AQUIFER SYSTEM

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Abstract

Nitrate concentrations in many of the public supply wells in the Northport area either approach or exceed the drinking water standard. At present, most land use in the area is residential, however, in the past, the area was extensively farmed. In order to determine what actions could be taken to reduce nitrate contamination in this part of the aquifer system, it is important to assess to what extent the nitrate contamination in this area is due to present land use activities as opposed to past agricultural activities. Residential contributions are believed to be composed of a combination of nitrates derived from turf grass fertilization and septic system wastes. Eleven samples were collected from the public supply wells in this area, one from each well field presently in operation. Another eleven samples were collected from shallow monitoring wells in the area. The geochemistry of the public supply well samples was compared to that of the monitoring well samples and other geochemical data from the literature.

The nitrogen and oxygen isotopes of groundwater nitrates were used in conjunction with other geochemical data to place constraints on the potential sources of nitrate. The oxygen isotopic composition of the nitrates in the public supply wells indicates that these nitrates are derived from nitrification of ammonia. Atmospheric nitrates and nitrate fertilizers are not an important source of nitrate in this area. In addition, the oxygen data suggests that denitrification is not an important process in this part of the aquifer. Dissolved oxygen measurements support this conclusion. Anaerobic conditions are required for denitrification to proceed and the lowest dissolved oxygen measured in any of the public supply wells was 6 mg/l. The severity of the nitrate contamination in this part of the aquifer system is probably due to the fact that the entire flow path is aerobic. In addition, the great depth to the water table may limit the amount of denitrification that occurs in septic system plumes resulting in a greater than average contribution of nitrates from this source.

Nitrate contamination in the supply wells in the Northport area appears to be linked to present rather than past land use activities. The public supply wells with the heaviest nitrogen isotopic composition (Waterside Road and Middleville Road) are clearly impacted by septic system wastes. Land use in the vicinity of the Waterside well is residential and the Middleville well may be influenced by onsite sewage treatment at the Veteran's Administration hospital. These are among the shallowest wells in the study area and would be those most likely to be affected by recent land use activities. The geochemistry of the Gun Club Road wells indicates that the nitrate in these wells is probably derived from agricultural activities. Current land use in the vicinity of these wells is agricultural. Several of the wells in the area have isotopic compositions that fall on the border between the agricultural and the residential fields. The water in most of these wells is geochemically more similar to that found in residential areas, however interpretation of the data is still in progress.

Introduction

The widespread contamination of groundwater with nitrate is of global concern to both environmentalists and public health officials. Discharge of contaminated groundwater into shallow marine bays contributes to the eutrophication of coastal waters and the decline of local fisheries. Ingestion of water which contains elevated concentrations of nitrate interferes with the ability of the blood to transport oxygen leading to the development of methemoglobinemia, a condition that is potentially fatal particularly for infants and small children. Therefore, the United States and Canada have set a drinking water limit of 45 mg/l as nitrate or 10 mg/l as nitrogen (Freeze and Cherry, 1979). The World Health Organization has recommended that similar standards be adopted worldwide (WHO, 1984).

The Long Island aquifer system presents a unique challenge to water resource managers because almost 7 million people 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 (Kreitler et al., 1978; Perlumutter and Koch, 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. Much of Long Island was once intensively farmed and to this day Suffolk County, on the eastern end of Long Island, remains one of the leading agricultural counties in New York state. However, the importance of agriculture on Long Island has continued to decline as urbanization has spread eastward from New York City. In order 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 which are due to previous agricultural activities and those which are due to urbanization and present land use practices. The objective of this study is to place constraints on the relative contributions of various land use activities to nitrate contamination of the aquifer. The results of this study are expected to be broadly applicable since urbanization of agricultural lands can be expected to place similar pressures on the development of water resources worldwide.

Previous research has demonstrated that the nitrogen and oxygen isotopic composition of groundwater nitrates can be used to help distinguish between different sources of nitrate (Aravena et al., 1992; Durka et al. , 1994; Wassenaar, 1995). 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; Bottcher et al., 1990). It is essential to collect additional geochemical data in order to strengthen the interpretation of the isotopic data because the isotopic composition of the nitrate observed may be affected by fractionation or the exchange of nitrogen between various pools (Komor and Anderson, 1993).

In order to assess the impact of different types of land use on nitrate contamination of the aquifer, groundwater samples were collected from both monitoring wells and public supply wells. Monitoring wells are generally 6" or less in diameter and have short screens that are located 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 types of land use. 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 collected from these wells were used to determine the average concentration and isotopic composition of nitrates present within a cross-section of the aquifer. The isotopic composition of the public supply well samples was compared to that of the various sources in order to place constraints on the relative contributions of each nitrate source to nitrate contamination within the aquifer. Additional geochemical data was collected to aid in the interpretation of the isotopic data.

Study Area Description

The Northport study area is located along the north shore of Long Island (figure 1). At least one sample was collected from each of the public supply well fields currently in operation in the Northport area. Nitrate concentrations in many of these wells either approach or exceed the public drinking water standard. (Wells that exceed the standard are no longer in use). Monitoring well samples were primarily collected from residential areas in Northport and neighboring Huntington (figure 1). 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.

Currently, land use is primarily residential (Long Island Regional Planning Board, 1982). Population density ranges from low (1 dwelling or less/acre) to intermediate (5-10 dwellings/acre). A small network of sewers exists in downtown Northport, however, most wastewater is disposed of through individual septic systems. Lawns are well maintained. Residential groundwater is expected to contain a mix of nitrates derived from fertilizers and septic system wastes. In the past, much of the area was intensively farmed. Older water in some of the deeper wells may contain nitrates derived from previous agricultural activities.

Hydrogeology

The hydrogeology of Long Island has been described in detail elsewhere (Jensen and Soren, 1971; Franke and McClymonds, 1972; Smolensky et al., 1989) and is only reviewed briefly here. 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, the Raritan confining unit, and the Magothy aquifer are Cretaceous Atlantic Coastal Plain deposits. The Upper Glacial aquifer consists of Pleistocene morainal deposits, outwash, and marine and lacustrine clays (Lubke, 1964). The groundwater divide is located in the late Wisconsin Ronkonkoma Moraine. Water flows north from the recharge area in the Ronkonkoma Moraine and discharges into Long Island Sound. To the south, it empties into the Atlantic Ocean.

The Northport study area is located to the north of the groundwater divide. The Upper Glacial aquifer rests unconformably on the irregular erosion surface of the Magothy except along the north shore where the Magothy has been completely scoured away. The Upper Glacial aquifer and the Magothy are hydraulically interconnected and it can be difficult to distinguish between the two since the Pleistocene glacial deposits are derived in large part from the underlying Cretaceous. Locally, the Smithtown clay acts as a confining layer within the Upper Glacial aquifer (Krulikas and Koszalka, 1983). Discontinuous thick layers of clay found in some of the wells in the northern part of the study area may be correlative with the Port Washington Confining Unit described in previous USGS studies of the stratigraphy of the north shore of Long Island (Kilburn, 1979; Kilburn and Krulikas, 1987).

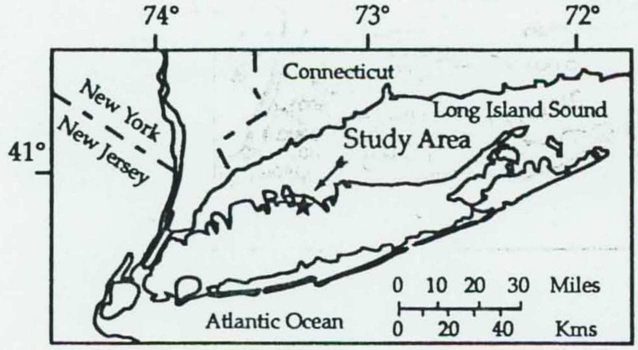

Figure 1A: Location Map

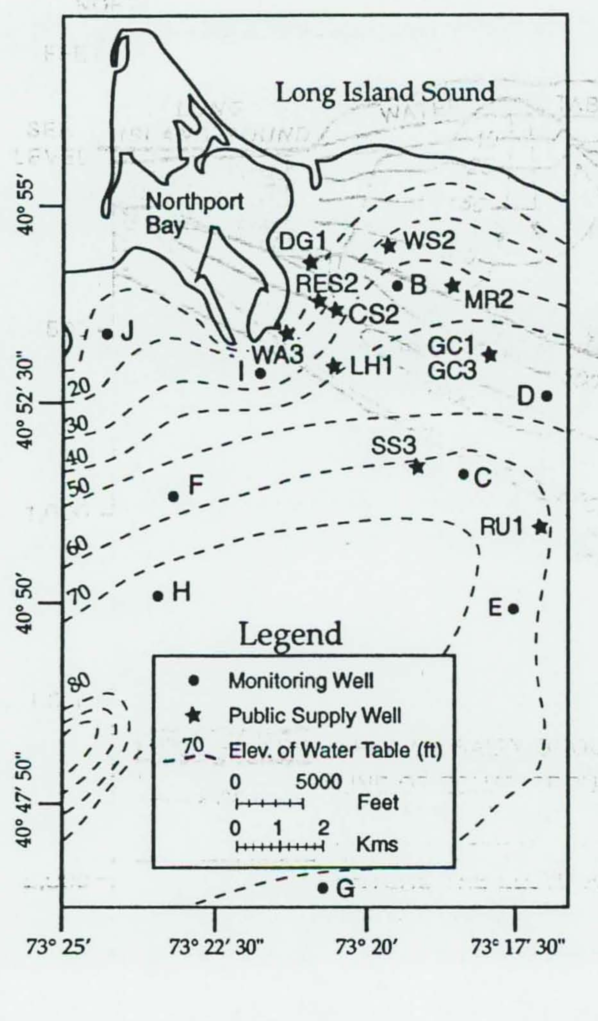

Figure 1B: Sketch Map of wells. March 1997 water table elevations provided by Suffolk County Department of Health Services

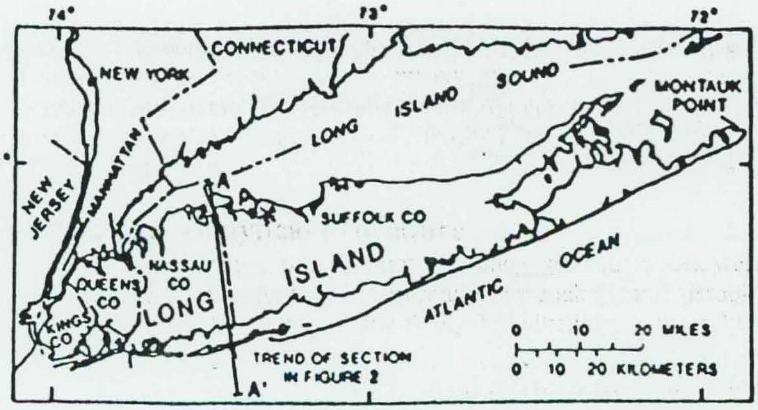

Figure 2a: Location of Cross Section modeled by Buxton and Modica (1992). The Northport study area is located approximately 15 miles east of the cross section line.

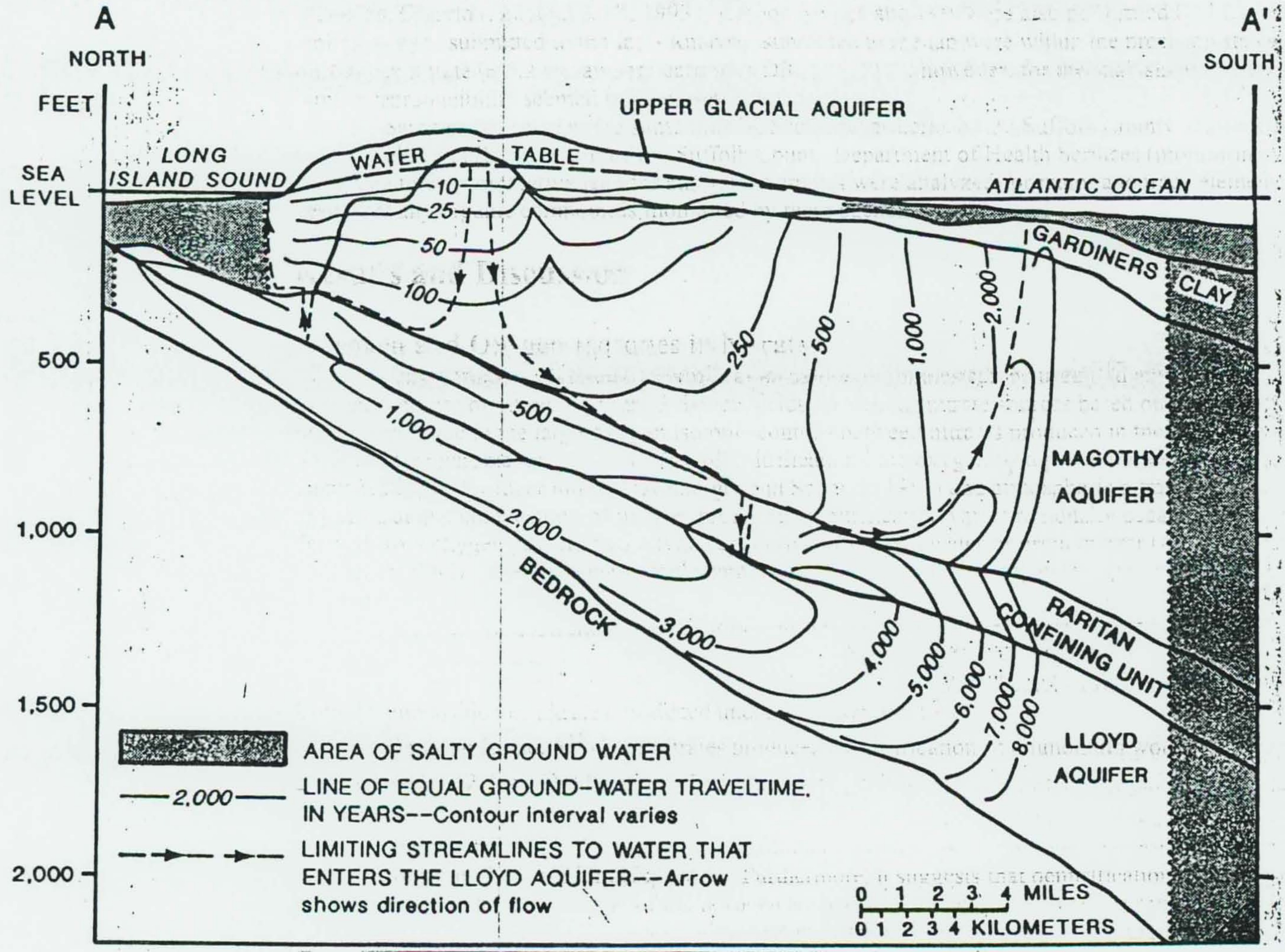

Figure 2b: Age of groundwater along modeled section (Buxton and Modica, 1992). The Northport study area lies to the north of the groundwater divide.

Groundwater flow has been modeled by the United States Geological Survey along a north/south transect near the Nassau/Suffolk border approximately 15 miles to the west (figure 2; Buxton and Modica, 1992). 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.

Field and Analytical Procedures

Temperature, pH, conductivity, and dissolved oxygen were measured in the field. Dissolved oxygen was also determined by Winkler titration on return to the lab at Stony Brook. The alkalinity of the monitoring well samples was determined by titration in the field. The alkalinity of the public supply well samples was determined by the Suffolk County Water Authority lab. 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 using a 0.45 micron flow-through filter 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 the nitrogen and oxygen isotopic composition of 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. Nitrogen and oxygen isotopic analyses were performed 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. Analyses submitted to the lab were within the precision stated by the lab for nitrogen in nitrate (± 0.2 ‰) and for carbon in DIC (± 0.1 ‰), however, for the analysis of oxygen in nitrate, natural sample reproducibility seemed to be closer to ± 1.0 ‰.

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 elements and the suite of pesticides and organic compounds monitored by these agencies

Results and Discussion

Nitrogen and Oxygen Isotopes in Nitrate

The nitrogen and oxygen isotopic composition of nitrates can be used to distinguish between different potential sources of nitrate. Figure 3 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 fertilizer nitrates (Amberger and Schmidt, 1987) and atmospheric nitrates (Durka, et al., 1994). The oxygen isotopic composition of nitrates produced by nitrification varies regionally because one oxygen in the nitrate is derived from oxygen gas and two oxygen are derived from soil water or groundwater (Anderson and Hooper, 1983; Hollocher, 1984). The nitrogen isotopic composition of these nitrates depends in part on that of the nitrogen source, however, this original signature may be modified by exchange with soil nitrogen pools or by processes such as ammonia volatilization (Kreitler, 1979; Flipse and Bonner, 1985) or denitrification (Mariotti, 1988; Bottcher et al., 1991) which depend upon local geochemical conditions. 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.

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 based on the isotopic composition of atmospheric oxygen ($\delta^{18}O$ = +23.5; Amberger and Schmidt, 1987) and that of Long Island groundwater ($\delta^{18}O$ = -7 to -8; USGS Watstore data). The oxygen isotopic composition data for the public supply wells sampled indicates that the nitrates in these wells are derived primarily from nitrification of ammonium (Figure 4). Furthermore, it suggests that denitrification, which results in a 2:1 enrichment in the heavier isotope of the nitrogen and oxygen isotopes respectively (Amberger and Schmidt, 1987; Bottcher 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 ranged from 58 to 100% saturated with respect to dissolved oxygen.

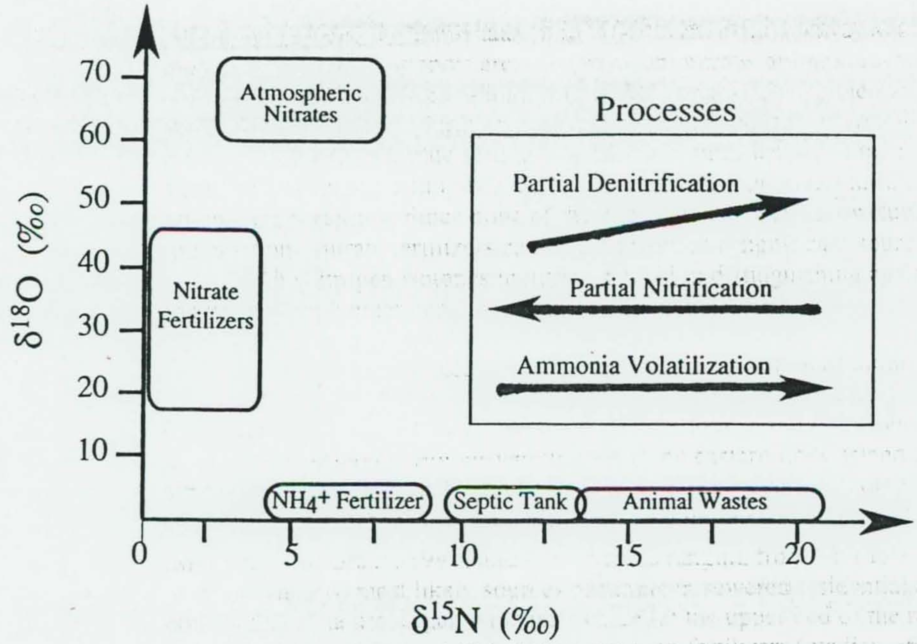

Figure 3: Nitrogen and oxygen isotopic composition of nitrate sources (Kreitler, 1975; Amberger and Schmidt, 1987; Bottcher et al., 1990; Aravena et al., 1993; Durka et al., 1994, Wassenaar, 1995)

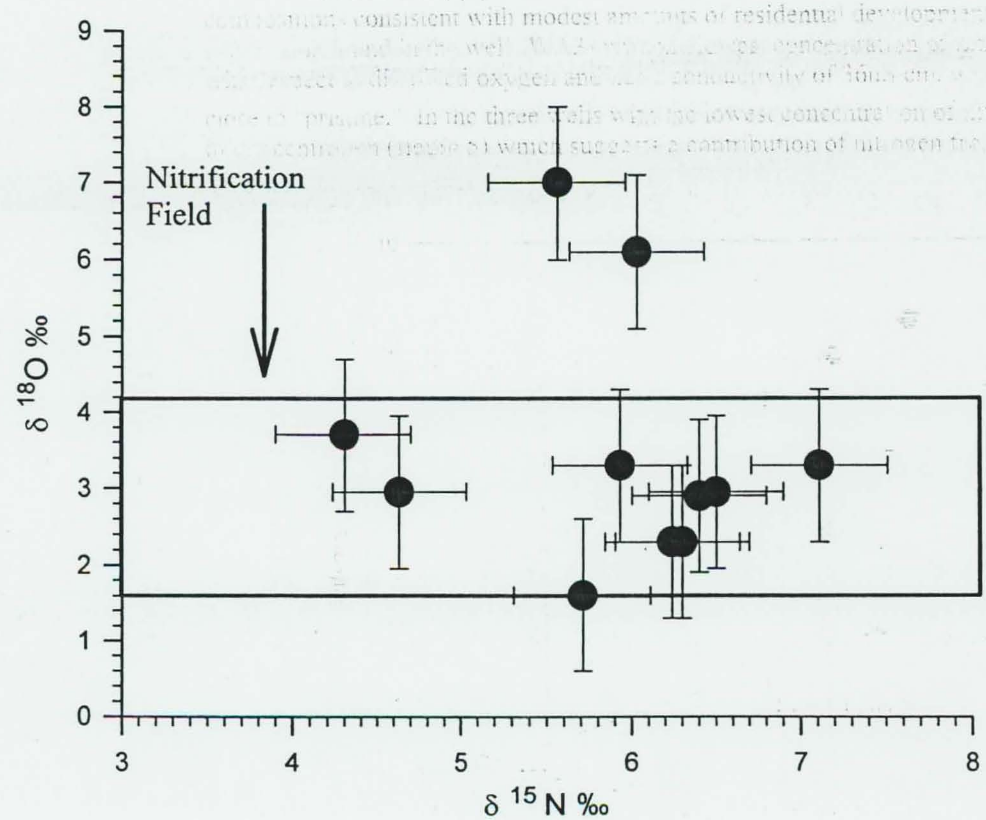

Figure 4: Nitrogen and oxygen isotopic composition of nitrates in public supply wells

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 the period 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 heavier oxygen isotopic composition. Since none of the public supply wells show substantially elevated oxygen isotopic compositions, nitrate fertilizers can be dismissed as a significant source of nitrate in this area.

The nitrogen isotopes in nitrate can aid in distinguishing agricultural from residential sources when used in conjunction with other geochemical data. Since the nitrogen isotopic composition of nitrates derived from agricultural activities has already been well-characterized for Long Island (Kreitler, 1978; Flipse and Bonner, 1985), this study focuses on characterizing the isotopic composition of nitrates associated with residential land use.

Nitrogen isotope values reported in the literature for sewered residential neighborhoods are similar to those reported for agricultural areas, but the concentrations of nitrate associated with these two types of land use are significantly different. For agricultural wells on eastern Long Island Kreitler (1978) reported a median nitrate-nitrogen concentration of 14 mg/l and a mean nitrogen isotopic composition of +5.3‰. Studies of sewered residential areas noted nitrogen concentrations ranging from 0.06 to 2.2 mg/l (Flipse et al., 1984) and 2.6 to 3.8 mg/l (Mullaney and Grady, 1997) and $\delta^{15}N$ values ranging from +1.1 to +7.1‰ and +4.3 to +6.1‰, respectively.

The two most likely sources of nitrate in sewered residential areas are ammonium fertilizers and the breakdown of natural organic matter. Values at the upper end of the range are consistent with those reported for nitrification of ammonium and organic nitrogen fertilizers (median +6.5‰ for a golf course, + 6.2‰ for potato fields; Flipse and Bonner, 1985). The lighter nitrogen isotopic compositions probably reflect a contribution of nitrate from the breakdown of organic matter. In this study, nitrates collected downgradient from the Smithtown waste transfer station had a nitrogen isotopic composition of +3.2‰. These nitrates are presumably derived from the breakdown of organic matter in the landfill.

The public supply wells with the lowest concentrations of nitrate have concentrations and nitrogen isotopic compositions consistent with modest amounts of residential development in upgradient areas. The lightest nitrogen (+4.3‰) is found in the well (WA3) with the lowest concentration of nitrate (0.88 mg N). This well is saturated with respect to dissolved oxygen and has a conductivity of 36uS/cm, which suggests that the water in this well is close to "pristine." In the three wells with the lowest concentration of nitrate, $\delta^{15}N$ increases linearly as a function of concentration (figure 5) which suggests a contribution of nitrogen from a heavier source to the wells at Laurel

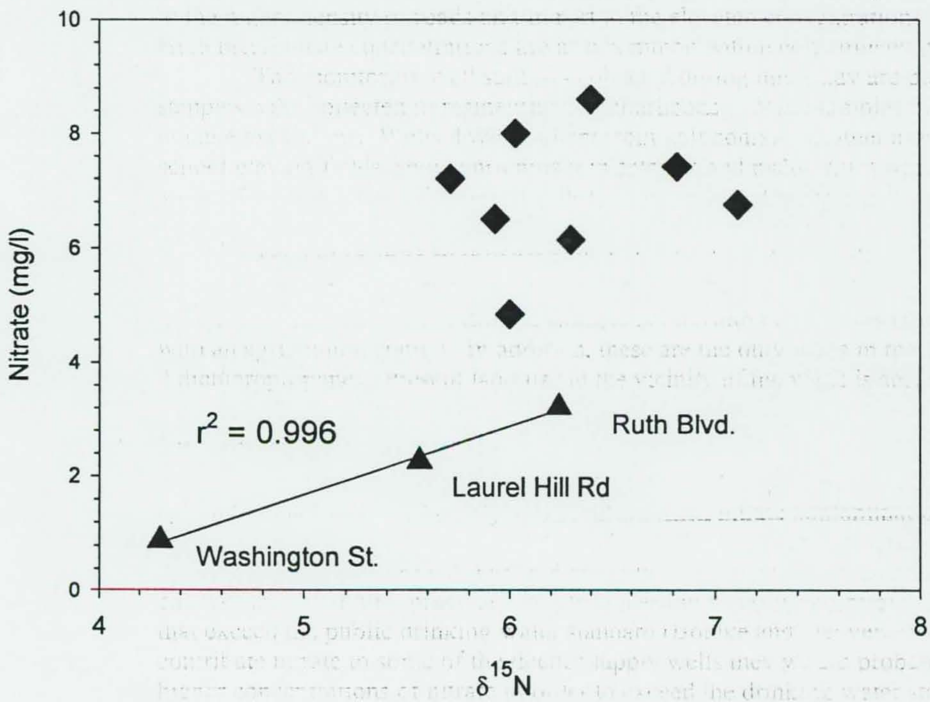

Figure 5: $\delta^{15}N$ vs nitrate concentration for public supply wells

Hill and Ruth Boulevard. From mass balance, $\delta^{15}N$ for the heavier source can be estimated to be +6.4 and +7.1‰, respectively. These values are similar to those observed for residential monitoring wells during this study.

The nitrogen isotopic composition of samples collected from residential monitoring wells ranges from +6.2 to + 8.4‰. Three of these wells are downgradient from areas of intensive turf grass cultivation such as golf courses (B, J) or playing fields (C). The others are suspected to be contaminated by septic system wastes (K, H, G). Nitrates in wells influenced primarily by turf grass cultivation are isotopically lighter (+6.2, +6.2, +6.5‰) than those influenced by septic system wastes (+6.7, +7.1, +8.4‰).

Older agricultural waters might also contain nitrates with heavier nitrogen isotopic compositions due to the application of manure to fields, however, additional geochemical data can be employed to 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 system, which subsequently leads to depletion in oxygen due to the respiration of organic carbon. In contrast, soil cultivation practices tend to produce more aerobic conditions. Residential samples show a positive correlation of $\delta^{15}N$ with sodium concentration (figure 6a) and a negative correlation with dissolved oxygen concentration (figure 7a). These same trends can be observed in public supply well samples (figures 6b, 7b). Therefore, this heavier nitrogen is probably associated with septic system wastes rather than fertilization of agricultural fields with manure.

The public supply wells with the most elevated nitrogen isotopic compositions are Waterside Road 2 (+7.11) and Middleville Road 2 (+6.82). At the Middleville Road site, chloride and sulfate concentrations have been reduced by approximately 25% by reinjection of system water over the winter. The nitrogen isotopic composition would likely have been higher before reinjection began. The shallower wells in both of these well fields have already been closed because the concentrations of nitrate exceed drinking water standards. The Middleville Road well field is downgradient from the Veteran's Administration hospital and may be impacted by onsite sewage treatment. The Waterside Road well field is downgradient from areas of medium to intermediate density residential development. These two well fields are among the shallowest well fields in the study area and would be those most likely to show the influence of recent land use practices.

Major Ion Chemistry of Agricultural and Residential Groundwater

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 (figure 8a). Agricultural land use produces 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 contaminant plumes.

The monitoring well samples collected during this study are plotted in figure 8b. 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 enrichment in calcium and magnesium which approaches that observed in agricultural areas. This is due to the liming of soils to increase pH. On the anion plot, an acidic waste contaminant plume with a high concentration of sulfate plots within the agricultural field.

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. These wells plot just outside the agricultural field on both ternary diagrams (figure 8c). The nitrogen isotopic composition of nitrates in theses 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.

Summary

It seems unlikely that the nitrate contamination problem in the Northport area is primarily due to previous agricultural activities. The only well field in which nitrate contamination can be clearly linked to intensive agricultural practices is that at Gun Club Road. Present land use in the vicinity of these wells is agricultural. Although it could be argued that "old agricultural waters" would be chemically and isotopically different due to changes in agricultural practices, less fertilizer-intensive agricultural practices do not produce concentrations of nitrate that exceed the public drinking water standard (Bohlke and Denver, 1995). Although "old agricultural waters" might contribute nitrate to some of the deeper supply wells they would probably have to be mixed with younger waters with higher concentrations of nitrate in order to exceed the drinking water standard.

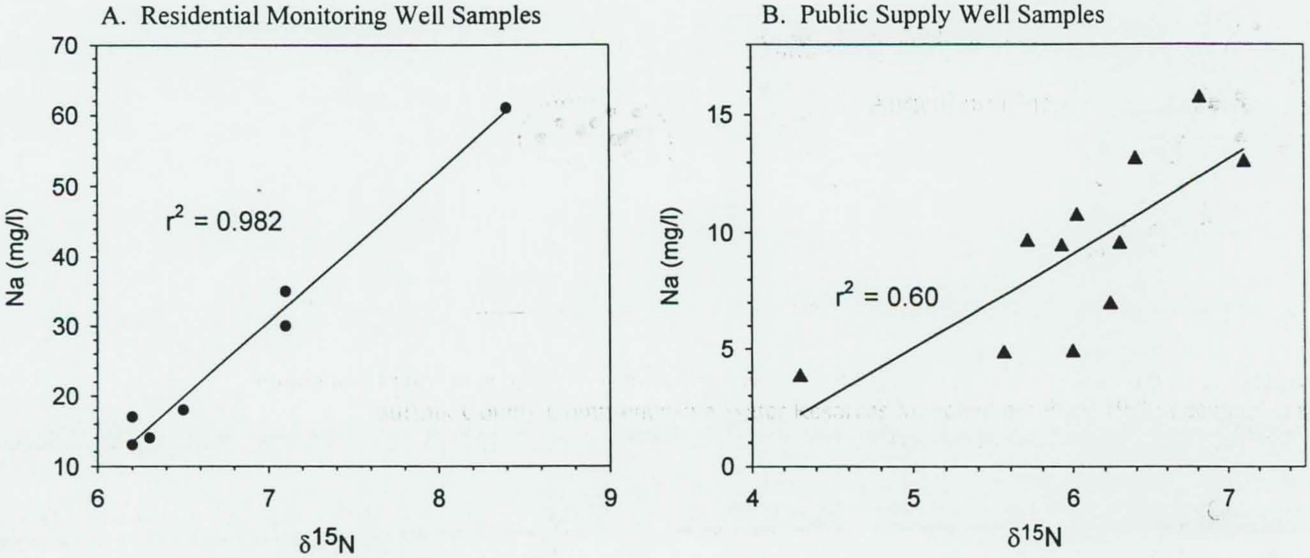

Figure 6: $\delta^{15}N$ vs Sodium Concentration

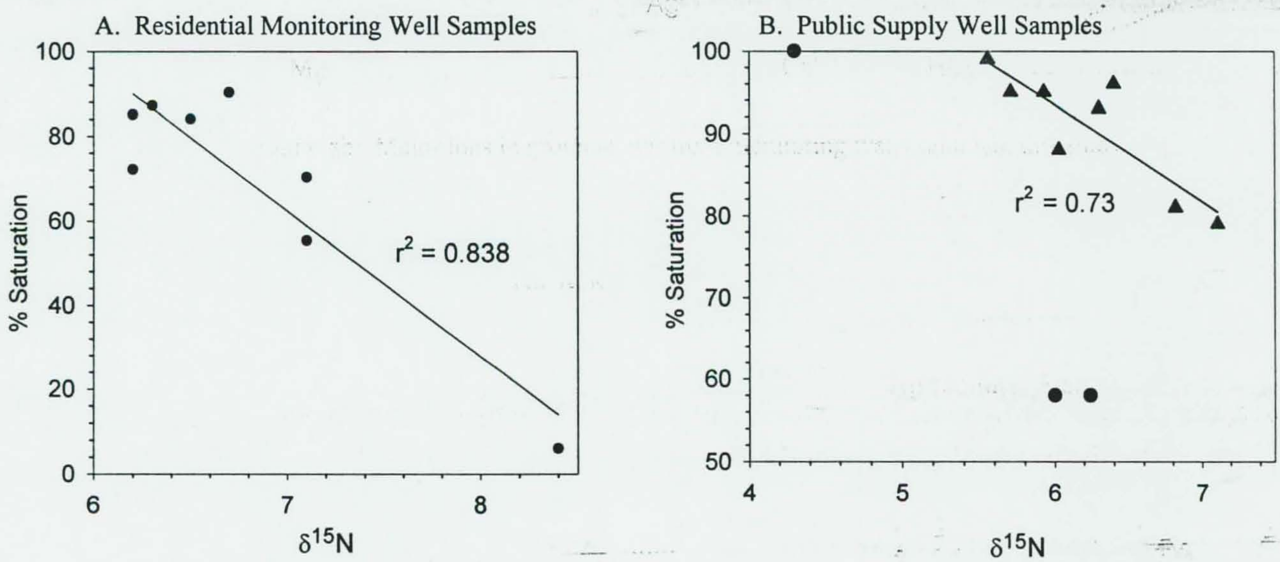

Figure 7: $\delta^{15}N$ vs Oxygen Saturation

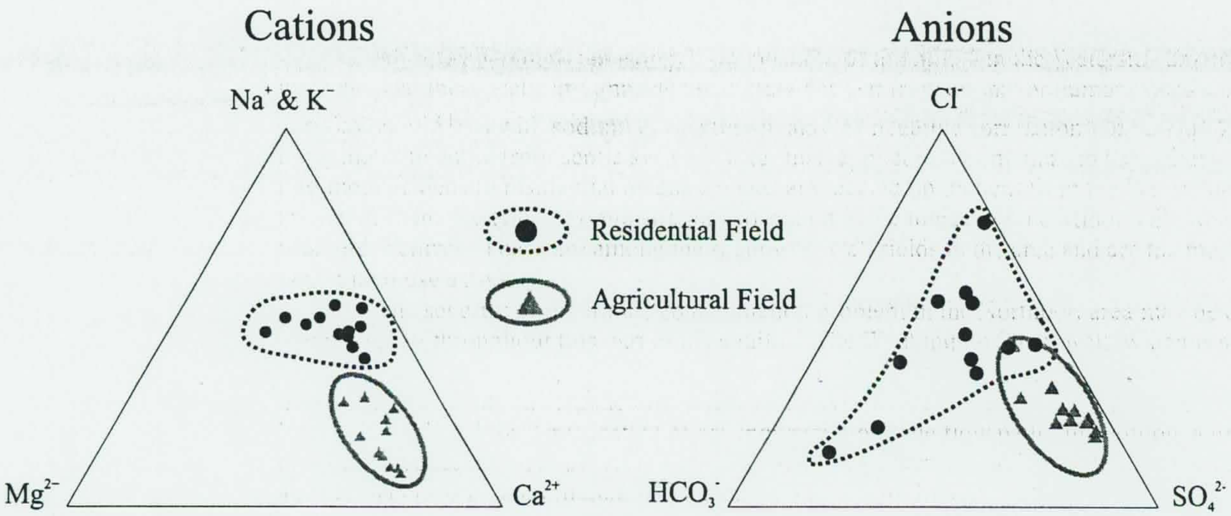

Figure 8a: Major Ions in Long Island Groundwater according to land use based on previous studies (deLaguna, 1964; Suffolk County Comprehensive Water Resorces Management Plan, 1987; Leamond et al., 1992; Pearsall, 1996)

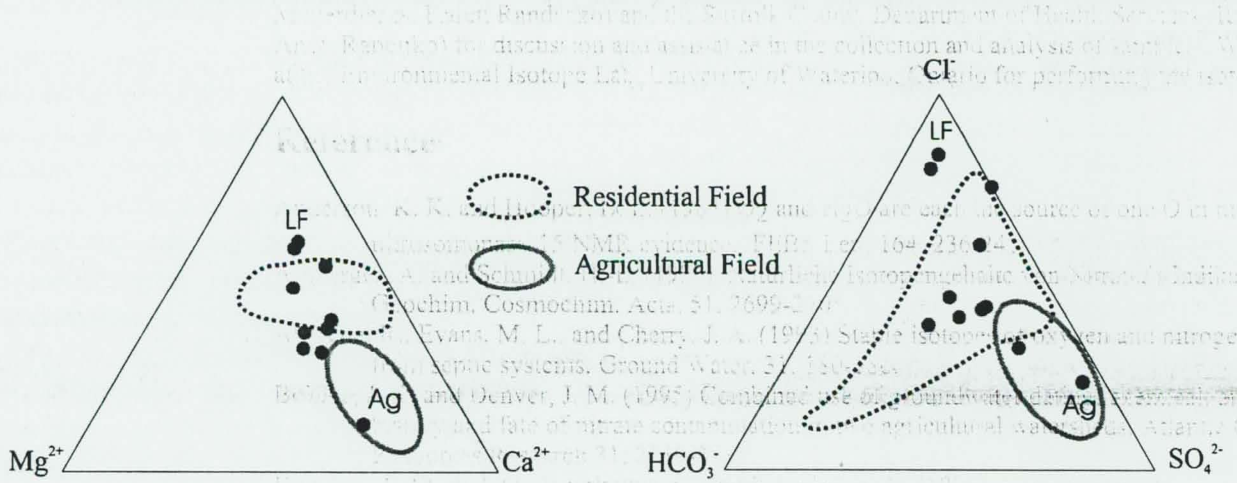

Figure 8b: Major ions in groundwater from monitoring wells sampled, this study.

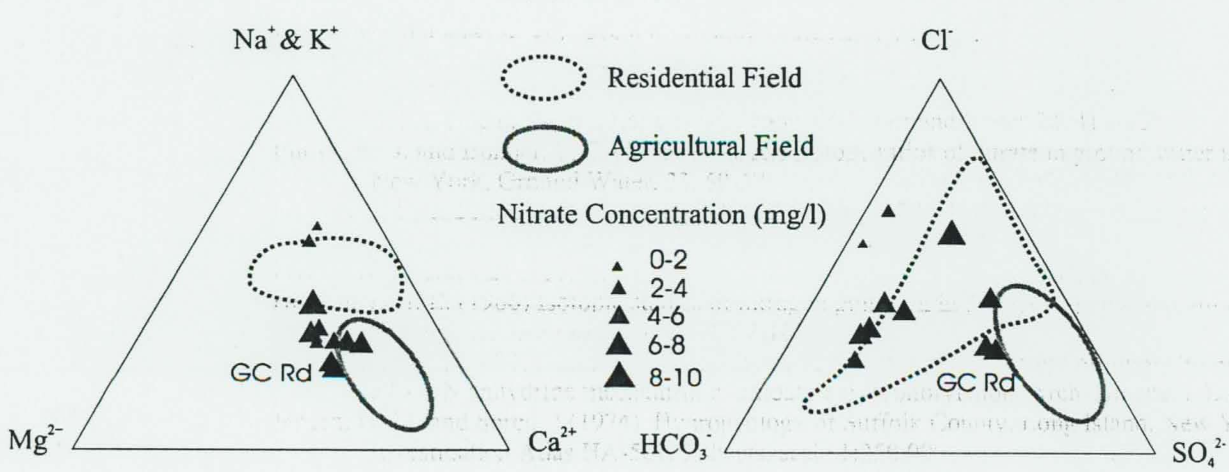

Figure 8c: Major ions in groundwater from public supply wells sampled, this study. Concentration of nitrate reflected by size of symbol.

The elevated nitrogen isotopic composition of the nitrate in the Waterside and the Middleville Road wells indicates that these wells are impacted by nitrate derived from animal or human wastes. Due to the positive correlation of $\delta^{15}N$ with sodium concentration and the negative correlation with dissolved oxygen, these nitrates are more likely to come from septic system wastes than application of manure to agricultural fields. Medium to intermediate density residential neighborhoods are located upgradient from the Waterside well. Onsite sewage treatment at the Veteran's Administration Hospital may be impacting 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 severity of the nitrate contamination problem in the Northport area may be due to the aerobic conditions, which prevail throughout this part of the aquifer. The Washington Street well, which is screened in the discharge zone, is still saturated with respect to dissolved oxygen. Although there are clays present in the Northport area that may contain organic carbon, the lowest dissolved oxygen recorded in any public supply well was 6 mg/l. No evidence has yet been found for denitrication of groundwater along the flow path. In addition, a greater than average proportion of the nitrogen present in septic plumes may enter the aquifer system due to the coarse gravelly nature of aquifer materials in combination with the great distance to the water table.

Acknowledgements

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