Occurrence of Nitrate in Groundwater—A Review

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ABSTRACT

The results of federal, state, and local surveys, which included more than 200 000 NO_3-N data points, are summarized in this review of NO_3 in groundwater in the USA. The levels of NO_3-N are associated with source availability and regional environmental factors. In regions where well-drained soils are dominated by irrigated cropland, there is a strong propensity toward the development of large areas with groundwater that exceeds the maximum contaminant level of 10 mg/L NO_3-N. Most of these areas are west of the Missouri River where irrigation is a necessity. Aquifers in highly agricultural areas in the southeastern USA reportedly are not contaminated. Vegetative uptake and denitrification in this warm, wet, C-rich environment are responsible for the natural remediation of NO_3 in shallow aquifers. In the Middle Atlantic states and the Delmarva Peninsula, localized contamination occurs beneath cropped, well-drained soils that receive excessive applications of manure and commercial fertilizer. Extensive tile drainage has for the most part prevented a NO_3 problem in the groundwater of the Corn Belt states. Throughout the USA there are recurring themes. They include a decrease in NO_3-N levels with depth; lower NO_3-N levels in shallow wells (< 8 m); and a significant increase in NO_3-N in older wells and in wells with poor construction. The factors affecting the distribution of NO_3 in aquifers are complex and poorly understood. Interdisciplinary studies using discrete depth sampling, hydrogeological indicators, isotopic tracers, and microbiological techniques are necessary to unravel the complex dynamics.

INGESTION of NO_3 in drinking water has caused methemoglobinemia in infants under 6 mo of age and recently caused the death of a South Dakota infant (Johnson et al., 1987). The noncancerous acute toxicity of NO_3 is the U.S. Environmental Protection Agency's (USEPA) basis for establishing a maximum contaminant level (MCL) for NO_3-N in drinking water. Although acute toxicity generally has been documented at concentrations greater than 50 mg/L NO_3-N, the MCL has been set at 10 mg/L. The documentation for the MCL recently was reviewed during a reevaluation of the reference dose (RfD). The USEPA's findings reaffirmed the 1.6 mg/kg per day RfD, which supports the present MCL of 10 mg/L NO_3-N (Dourson et al., 1991). In rural community water systems where NO_3 levels exceed the MCL, the health concerns for this segment of the population have been protected by providing an alternative source of drinking water. Because the present technologies for removal of NO_3 from drinking water have serious limitations that make their application difficult to prescribe (Dahab, 1991), many communities have had to find new sources of water. Whether that source is surface water or groundwater from a deeper formation, it is an economic hardship for these small communities to bring their water supply into compliance with the NO_3 provision of the Safe Drinking Water Act.

Additional research is necessary to confirm the relationships between ingestion of NO_3 in drinking water and hypertension (Malberg et al., 1978), increased infant mortality (Super et al., 1981), central nervous system birth defects (Dorsch et al., 1984), and certain cancers including stomach cancer (Hill et al., 1973) and non-Hodgkin's lymphoma (Weisenburger, 1991). Neither the noncarcinogenic health effects of NO_3 nor the carcinogenic effect of NO_3 or N-nitroso compounds, produced when NO_2 formed by bacterial reduction of NO_3 reacts with nitrosatable substrates, have been proven conclusively. The experts (Weisenburger et al., 1991) agree that the majority of evidence implicating NO_3 as a causative factor is based on correlation studies that provide only weak evidence of an association and should not be used to establish a cause-and-effect relationship.

Historically, the justification for surveys of NO_3 levels in groundwater and for research into NO_3 behavior in groundwater has been the adverse health effects attributed to the ingestion of NO_3 in drinking water. Now, however, there is growing environmental awareness that NO_3 is the limiting nutrient in nearshore environments and in several lakes (Ryther and Dunstan, 1971; Viner and White, 1987). Concerns for nutrient levels in surface waters are fueling NO_3 studies in the Chesapeake Bay (Glibert et al., 1991), the Mississippi Delta (Turner and Rabalais, 1991), and the Aegean Sea (Ganoulis, 1991). Because groundwater is the major component of baseflow in creeks and rivers, an understanding of its contribution to the NO_3 load in surface water is important. As a result there is a renewed effort to protect streams from shallow groundwater inputs of NO_3 as well as NO_3, suspended sediments, P, and pesticides in storm runoff. Under certain flow and redox conditions, riparian zones along waterways have demonstrated a natural proficiency to intercept and denitrify NO_3 in shallow groundwater (Gilliam, 1991; Schipper et al., 1991).

NITRATE IN GROUNDWATER

Global Synopsis

Nitrate is the most ubiquitous chemical contaminant in the world's aquifers and the levels of contamination are increasing. Increased degradation of drinking water and eutrophication of coastal waters in the European Community (EC) are consequences of steadily increasing NO_3 levels in surface, ground, and coastal waters (Fried, 1991). In areas of Belgium where agriculture has adversely affected groundwater quality, NO_3-N concentrations range from ≈4 to 11 mg/L. By 1995 more than 10 million French, 20% of the population, will drink water exceeding the EC limit of 11.3 mg/L NO_3-N. Eight percent of the public waterworks in Denmark and 5% of those in the former Federal Republic of Germany supply groundwater that exceeds the EC limit for NO_3-N and in both countries unprotected aquifers are contaminated with

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Abbreviations: USEPA, U.S. Environmental Protection Agency; MCL, maximum contaminant level; RFD, reference dose; EC, European Community; WATSTORE, Water Storage and Retrieval System; NAWWS, National Alachlor Well Water Survey; NPS, National Pesticide Survey.

NO_3 (Fried, 1991). Increasing NO_3 levels in the groundwater of the Netherlands eventually will effect 25% of the wellfields (Fried, 1991). Nitrate levels in many private wells in these three countries are unacceptable. In eastern and central England, NO_3 concentrations are increasing in the groundwater, which provides 30 to 50% of the drinking water (Chilton and Foster, 1991). If current inputs of N from agricultural land continue, NO_3 levels in the groundwater will be double the EC limit. Nitrate in the groundwater of several agricultural areas in southern Ontario, Canada, also exceeds 10 mg/L NO_3-N (Gillham, 1991).

Nitrate contamination of groundwater is also a growing problem in the Caribbean, Africa, the Middle East, Australia, and New Zealand. Agriculture has affected groundwater quality in Barbados' two major catchments (Chilton, 1991). Although average NO_3-N concentrations in the rural areas remained below the 10 mg/L World Health Organization guideline, nowhere in the catchments were NO_3 concentrations indicative of a pristine environment. Concentrations in the urban areas were consistently above 10 mg/L NO_3-N and reflect N loading from high density housing with unsewered sanitation. Faillat (1990) reported NO_3 contamination of groundwater beneath areas of the Ivory Coast that were deforested and used for either crop production or settlements. Sewage effluent is an important source of water for agriculture in Israel and other semiarid areas with limited ground and surface water. In Israel, NO_3 contamination of shallow groundwater beneath sewage-irrigated land was attributed to applications of fertilizer and sewage effluent (Ronen and Magaritz, 1985). Nitrate-N levels in one-third of the bores sampled on the Gambier Plain in southeast South Australia exceeded 10 mg/L (Dillon et al., 1991). Leachates from leguminous pastures grazed by livestock were the source of most of the NO_3 in the groundwater, whereas NO_3 in wastes from dairies, saleyards, and milk and meat processing facilities was responsible for the highest concentrations. In New Zealand unconfined aquifers often are extensively contaminated by NO_3 (Burden, 1982). The contamination is associated with intensive grazing on nonirrigated grass–clover pasture, grazing on irrigated pasture, and fertilization of crop and horticultural land.

Surveys in the United States

Madison and Brunett's (1985) mapping of NO_3 concentrations in more than 87 000 wells was the first comprehensive, nation-wide evaluation of the areal distribution of NO_3 in groundwater (Fig. 1). Their database was the 25-yr record of NO_3 analyses in the U. S. Geological Survey's Water Storage and Retrieval System (WATSTORE). Although the data, collected largely from special project areas and municipal wells, are not temporally, regionally, or vertically representative (Madison and Brunett, 1985), they are useful for identifying regions with NO_3 problems. Nitrate-N concentrations exceeded 3 mg/L, Madison and Brunett's background level for NO_3-N in U.S. aquifers, in agricultural areas of Maine, Delaware, Pennsylvania, central Minnesota, Wisconsin, western and northeastern Iowa, the Plains states of Texas, Oklahoma, Kansas, Nebraska, and South Dakota, eastern Colorado, southeastern Washington, Arizona, and central and southern California. Lee and Nielsen (1989) used Madison and Brunett's data together with information on aquifer vulnerability and N fertilizer usage to delineate areas where there is a potential for NO_3 contamination of the groundwater. These refinements led to the elimination of areas with elevated NO_3-N concentrations in northern Maine and to the inclusion of areas in Ohio, Indiana, and Illinois where WATSTORE data were sparse. The studies of Madison and Brunett (1985) and Lee and Nielsen (1989) showed that nationally the presence and the predicted occurrence of NO_3 in groundwater are nonuniform and are skewed toward the central and western USA. The 3 mg/L NO_3-N background level can be debated as being too conservative from an environmental perspective, because half the samples collected across the USA do not have detectable levels of NO_3 (USEPA, 1990). From a regulatory standpoint, however, it is more practical to delineate areas with higher NO_3 levels such as those that exceed the MCL.

An objective of both Monsanto Company's (1990) National Alachlor Well Water Survey (NAWWS), and USEPA's (1990) National Pesticide Suyrvey (NPS) was to estimate with statistical accuracy the proportion of the population served by drinking water wells that exceed the nitrate MCL. In the NAWWS 1430 domestic rural wells in 89 counties in 26 states were sampled, whereas 783 rural domestic wells in 90 counties in 38 states and 566 community water systems in 50 states were sampled in the NPS. There are ≈10.5 million rural domestic wells and ≈ 38 300 community water systems in the USA. In the NPS, the estimated incidence of contamination, defined in this paper as NO_3-N levels in excess of the 10 mg/L MCL, was lower in the high capacity community water systems (1.2%) than in the domestic wells (2.4%) (Fig. 2). This is to be expected. High capacity wells pump groundwater from a larger vertical interval of the saturated zone than do domestic wells; consequently, concentrations of contaminants, which usually are higher at the top of the aquifer, are diluted. The discrepancies in the results for the rural domestic wells in the two surveys (Fig. 2) are related to land use. Because sampling sites in the NPS were selected without regard to pesticide or land use, inclusion of nonagricultural areas significantly lowered the number of wells that exceeded the MCL. The NAWWS, on the contrary, was restricted to regions of alachlor use. Because these are areas that are intensively cropped to corn (*Zea mays* L.) and soybean [*Glycine max* (L.) Merr.], N fertilizer, livestock wastes, and mineralized N from legumes and other organic matter are potential sources of NO_3 contamination. Consequently, the results of the NAWWS are a "worst-case" estimate of the incidence of NO_3-contaminated wells. From the NPS, USEPA (1992) estimates 4.5 million people including 66 000 infants under 1 yr of age are served by community water systems or rural domestic wells that exceed the 10 mg/L NO_3-N MCL.

In large-scale studies like the NPS, the relatively small sample population in comparison to the total study area provides only minimal insight into the factors affecting the occurrence and distribution of NO_3. Many of the weak associations between NO_3 concentrations and land use and well construction identified in the NPS (USEPA, 1992) have been thoroughly documented in smaller-scale investigations. Parallel conclusions reached by the NPS and state and county surveys do, however, demonstrate

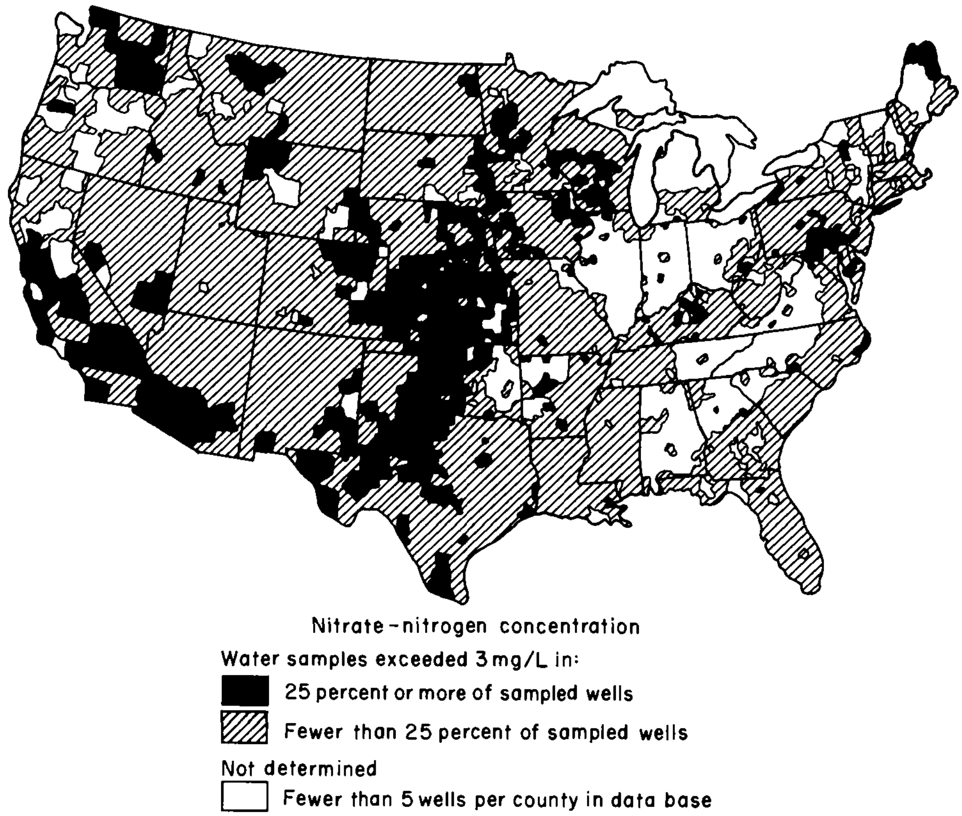

Fig. 1. Areal distribution of NO_3-N concentrations in groundwater in the contiguous USA (Madison and Brunett, 1985).

that the findings of the latter are substantiated on a national scale.

State-wide surveys of NO_3 concentrations in rural wells have been undertaken in Iowa (Kross et al., 1990), Kansas (Steichen et al., 1988), Nebraska (Spalding, 1991), North Carolina (Jennings, 1992, unpublished data), Ohio (Baker et al., 1989), and Texas (TSSWCB, 1991). Although sampling sites in the North Carolina, Ohio, and Texas surveys were not selected by a statistically randomized design, the sheer number of samples in each study negates most of the statistical bias introduced with a smaller sample. Nine thousand wells were sampled in the North Carolina survey, 16 166 in the Ohio study, and 55 495 in the Texas survey. Figure 3 summarizes the incidence of NO_3 contamination in these surveys. The highest incidence of contamination occurs in groundwater in the middle of the contiguous USA where NO_3 levels in $\approx$20% or more of the sampled wells in Iowa, Nebraska, and Kansas exceed the MCL. In contrast, the incidence of contamination is lower in Texas (8.2%) and very low in North Carolina (3.2%) and Ohio (2.7%).

Known or suspected NO_3 contamination prompted intensive surveys encompassing several counties in Arkan-

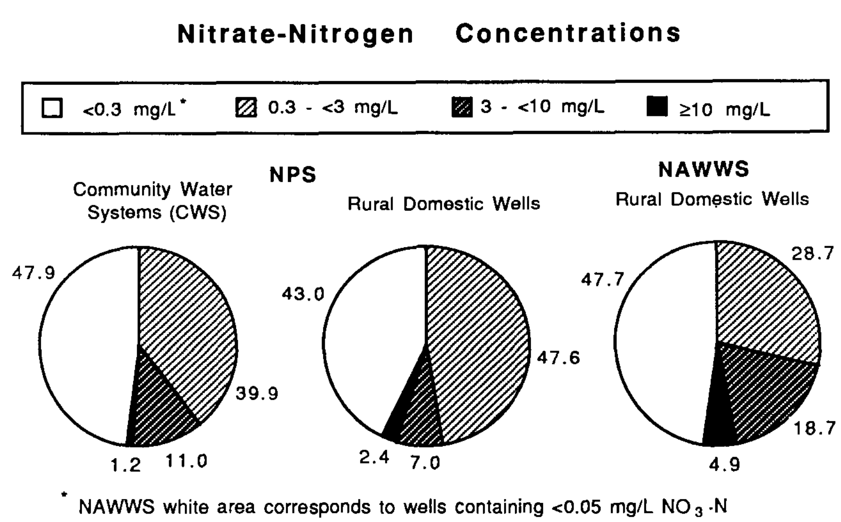

Fig. 2. Incidence of NO_3-N contamination in the National Pesticide Survey (USEPA, 1990) and the National Alachlor Water Well Survey (Monsanto, 1990).

sas (Arkansas CES, 1990), California (Anton et al., 1988), Delaware (Ritter and Chirnside, 1984), Pennsylvania (Pionke and Gilmeister, 1991), Washington (Erickson and Norton, 1990), Minnesota (Ruhl, 1987), and South Dakota (Goodman, 1985). The incidence of contamination reported in these studies is shown in Fig. 3. In the northeastern USA, NO_3 contamination occurred frequently in the agricultural areas of Pennsylvania and Delaware. Ten percent of the sampled wells in Pennsylvania's 10 most agriculturally based counties exceeded the MCL (Pionke and Gilmeister, 1991). Excessive leachate from field application of animal wastes and fertilizers caused Lancaster County to have some of the worst NO_3 contamination in Pennsylvania. Well-drained soils and the presence of several NO_3 sources cause the groundwater in recharge areas of the Delmarva Peninsula to be vulnerable to NO_3 contamination (Ritter and Chirnside, 1984). The highest incidence of contamination was in the intensive broiler producing area of coastal Sussex County where 37% of the wells exceeded the MCL. Leachates from poultry manure appeared to be the major contributor of NO_3 to the groundwater in four of the five problem areas. Leachates from commercial fertilizer applications and septic systems were secondary sources of NO_3 contamination.

The results from intensive state-wide monitoring and smaller studies that have used a variety of tracers offer opportunities to understand the prevalence of NO_3 contamination in the groundwater of some regions of the USA rather than others. The intensive groundwater NO_3 surveys recently completed in North Carolina, Ohio, and Nebraska, and ongoing groundwater research programs in these states are focused on understanding the management, distribution, and persistence of NO_3 in groundwater. Because these states represent three different regions of the USA, the results of these investigations are stressed in this report. Data from surrounding states will be introduced to describe other factors that may be representative on a regional scale. It is recognized, however, that there are areas within these large regions that do not fit the generalities. Additional information from other states can be found in Power and Schepers (1989), Fedkiw (1991), and Spalding and Exner (1991).

North Carolina and the Southeast

Results from the 3-year North Carolina Statewide Nitrate Survey of 9000 wells indicated that NO_3 contamination is neither a widespread nor a severe problem (Jennings, 1992, unpublished data). Well depth was a dominant factor affecting NO_3 concentrations. Besides shallow well depths, substandard well construction and the siting of wells near potential sources of contamination also were associated with higher NO_3 concentrations. Although the majority of the wells sampled in the survey were from the intensively farmed Piedmont Plateau and the Coastal Plain, the incidence of contamination was only 3.2%. The obvious question then is "Why is the incidence of contamination so low?"

Despite a 400% increase in fertilizer usage in North Carolina since 1945, NO_3 levels in streams have not changed (Jacobs and Gilliam, 1985). An analysis of 20 yr of data collected by the Soil Science Department at North Carolina State University led Gilliam (1991) to conclude that NO_3 is not a problem in groundwater downgradient from properly fertilized fields. He, however, qualified this statement by warning that N applications above recommended rates have caused documented increases in groundwater NO_3. Gilliam (1991) also observed that NO_3 levels of 15 to 20 mg/L NO_3-N occurred only in soil water beneath fertilized cornfields on the Coastal Plain and were never encountered below 4 m. Gilliam and his colleagues have determined that sufficient dissolved organic matter percolates from the high organic matter Coastal Plain soils to the shallow groundwater to provide the electrons for microbial reduction of NO_3 and that denitrification is largely responsible for the loss of most of the NO_3 from agricultural land in the Coastal Plain (Daniels et al., 1975; Gambrell et al., 1975; Jacobs and Gilliam, 1985). In highly instrumented studies, reductions in NO_3 levels also have been documented in shallow groundwater beneath riparian forests in Maryland (Peterjohn and Correll, 1984; Jordan et al., 1993) and Georgia (Lowrance and Pionke, 1989; Lowrance, 1992). The reduction dynamics are complex and can result from either vegetative uptake, denitrification, or a combination of the two.

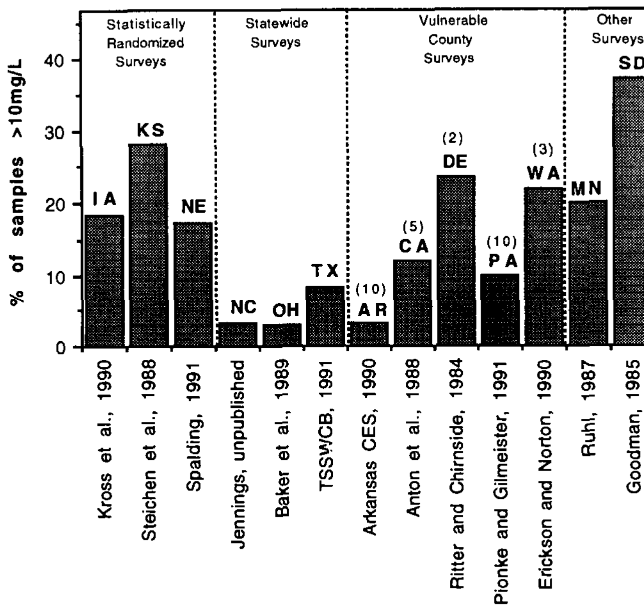

Fig. 3. Incidence of NO_3-N contamination in large selected surveys (number in parentheses is number of counties surveyed).

Hubbard and Sheridan (1989) reported that despite heavy applications of fertilizer, abundant precipitation ($\approx$127 cm/yr), and rapid irrigation development of the sandy soils in the Piedmont and Coastal Plain of the southeastern USA, there are few instances of aquifer NO_3 contamination. In a 10-county pilot study in Arkansas, only 3.2% of the 1232 wells sampled had NO_3 levels above the MCL, whereas 86% of the sampled wells had concentrations less than 3 mg/L NO_3-N (Arkansas CES, 1990). High NO_3 concentrations frequently were encountered in areas of intensive poultry and livestock production. Steele and McCalister (1991) reported that even in areas receiving heavy applications of N from poultry litter, NO_3 concentrations in the groundwater averaged <3 mg/L. It appears that the high temperatures, abundant rainfall, and the relatively high organic content soils in the Piedmont Plateau and Coastal Plain of the southeastern USA promote denitrification below the root zone and naturally remediate NO_3 loading of the groundwater.

Ohio and the Corn Belt

The Corn Belt states of Iowa, Missouri, Illinois, Indiana, and Ohio have some of the most productive and intensively cropped land in the world. The region is characterized by flat, fertile land; hot, humid summers; and ample rainfall. The Corn Belt has both the largest proportion of cropland (58%) and the largest proportion of corn cropland (30%) in the USA (U.S. Dep. of Commerce, 1989).

Although Lee and Nielsen's (1989) vulnerability assessment showed that groundwater in Ohio is vulnerable to NO_3 contamination from agriculture, concentrations exceeded the MCL in only 2.7% of the 14 478 domestic wells sampled in Baker et al.'s (1989) state-wide survey and exceeded 3 mg/L NO_3-N in only 12.7% of the wells. The average concentration was 1.3 mg/L NO_3-N. Given the low levels of NO_3, Baker et al. (1989) assumed that concentrations less than 0.2 mg/L NO_3-N represented natural background levels in the groundwater.

Documentation of numerous siting and construction variables that could be associated with NO_3 contamination in wells was an integral part of Baker et al.'s (1989) study. High levels of NO_3 occurred more often in older wells, shallow (<15 m) wells, and poorly constructed wells than in their counterparts. Poor construction refers to construction that is well below standards deemed acceptable today. Denitrification can be a factor in this stratification of NO_3 with depth. Decreases in NO_3 concentrations with increasing depth also have been reported in aquifers in Illinois (McKenna et al., 1988; Schock et al., 1992) and Iowa (Hallberg, 1989). The average NO_3 concentration was higher in wells located within 60 m of cropland and feedlots; however, this was not true for wells within 60 m of septic systems. Interestingly, although there was an overall increase in NO_3 levels in wells sited close to cropland, counties with the most intense cropping had some of the lowest average NO_3 levels.

The low incidence of NO_3 contamination, the low average concentrations, and the associations between concentration and land use in Ohio suggest that significant quantities of surface-applied N are not reaching the groundwater. Logan et al. (1980) showed that leachates from cropped land were intercepted by tile drains and discharged to surface waters. They measured losses of 20 to 100 kg NO_3-N/ha per yr from tile-drained fields at numerous sites in several North-Central states. These losses also explain the occurrence of NO_3-N in concentrations above the MCL in the Scioto River at Columbus, OH, during the spring. Power and Schepers (1989) hypothesized that the diversion of NO_3-contaminated recharge by tile drains and its subsequent discharge to surface water is the major fate of surface-applied N in several states in the eastern Corn Belt.

Iowa, the state with the highest N fertilizer use, appears to be a transition zone between relatively minor groundwater NO_3 problems in the Corn Belt states to the east and extensive NO_3 contamination in the groundwater of the Plains states. The results of Iowa's statistically representative State-Wide Rural Well-Water Survey (Kross et al., 1990) showed that the regional distribution of NO_3 concentrations in excess of the MCL was not uniform and was skewed. North central Iowa had the lowest incidence of contamination (5.6%). Contamination occurred more frequently in south-central Iowa where NO_3 levels exceeded the MCL in 28.1% of the wells, whereas the highest incidents of contamination were in the glaciated areas of southwest and northwest Iowa where 31.4 and 38.2%, respectively, of the wells exceeded the MCL. A major difference between the areas with high (>25%) and low (<25%) incidents of contamination was related to well depth and well construction. Most (63 – 73%) of the wells in northwest, southwest, and south-central Iowa are large diameter seepage wells that act as sumps for the collection of infiltrate. By design they are shallow wells without a watertight casing; consequently, they are very vulnerable to surface contamination a fact also borne out by the very high incidence (83%) of total coliform contamination (Kross et al., 1990). Because results from areas with large numbers of poorly constructed wells are not representative of aquifer conditions and tend to overestimate the problem, the contibution from nonpoint sources in these regions is difficult to assess.

Nebraska and the West

The topography and climate of Nebraska, centered in the Great Plains, cause agricultural practices to be quite different from those in the Corn Belt and the southeastern USA. Irrigation and an abundant supply of easily accessible groundwater enable Nebraska to irrigate about 2.3 million ha of cropland (U.S. Dep. of Commerce, 1989). Only California has more irrigated cropland (≈3.1 million ha). Nebraska is consistently one of the three leading corn-producing states; the others are Iowa and Illinois.

The impact of NO_3 from all potential sources appears more direct in the groundwater of Iowa, Nebraska, and the western states. Commercial fertilizer and mineralized N from both natural accumulations of organic matter and crop residues are dispersed sources of NO_3 that have contaminated groundwater. Both cultivation and irrigation, especially when excessive irrigation occurs on well-drained soils, exacerbate the vertical transport of NO_3 from these sources. Mineralized natural organic N is a source of NO_3 contamination in the Montana saline seep area (Miller et al., 1981) and a potential contaminant in southwestern Nebraska (Boyce et al., 1976). In the Sand Plain Aquifer region of Minnesota where 20% of the wells exceeded the MCL (Ruhl, 1987), the incidence of contamination in the irrigated cropland area approached 50%. Concentrations averaged 17 mg/L NO_3-N in the irrigated areas and 5.4 mg/L in the nonirrigated cultivated areas (Anderson, 1989). The sand plain region of central Wisconsin is another area where the groundwater is contaminated by leachates from irrigated agriculture (Saffigna and Keeney, 1977). Power and Schepers (1989) point to the association of irrigation with the large areal NO_3-N contaminated areas in the states of Arizona and California. Historically the groundwater beneath the intensely farmed and irrigated basins in central and southern California has had high concentrations of NO_3 (Ward, 1970). In six southern California counties NO_3 levels in ≈12% of the municipal wells exceeded the MCL (Anton et al., 1988). Primarily as a consequence of exceeding the MCL, ≈4% of the domestic groundwater supply is lost annually. In contrast, less than 0.5% of the supply has been lost to organic chemical contamination. Fertilizer leachates from lawns also are a potential threat to groundwater contamination (Exner et al., 1991). Goodman (1985) reported that livestock containment areas and accidental fertilizer releases were two point sources responsible for much of the NO_3 contamination in the Big Sioux aquifer in South Dakota. Poor well construction and improper siting of wells compound the contamination. In Washington, additional point sources of NO_3 contamination are lagoon seepage and illegal discharges in dry wells (Spalding et al., 1982). A full treatise of sources of NO_3 contamination is given in Keeney (1986).

A recent compilation (Exner and Spalding, 1990) of NO_3 data from 5826 wells sampled in Nebraska between 1984 and 1988 indicated that NO_3-N concentrations exceeded the MCL in more than 20% of the wells. Slightly more than half of the wells that exceeded the MCL were in areas highly vulnerable to leaching (Fig. 4). These areas are characterized by fence-row to fence-row irrigated corn grown on well to excessively well-drained soils and a vadose zone less than 15 m thick. Because much of the data was from site investigations, it is concentrated in areas with elevated NO_3 concentrations. The more than 202 000 contiguous hectares underlain by NO_3-contaminated groundwater in the central Platte region (Buffalo, Hall, and Merrick counties in Fig. 4) are the largest areal expanse of NO_3-contaminated groundwater in Nebraska. Nitrate levels in this groundwater have increased at rates of 0.4 to 1.0 mg NO_3-N/L per yr (Exner, 1985). Studies measuring NO_3 and pesticide concentrations and N stable isotope ratios in groundwater from both existing wells (Exner and Spalding, 1976; Gormly and Spalding, 1979; Spalding et al., 1979, 1980) and upgradient and downgradient nested monitoring wells (Junk et al., 1980; Spalding and Exner, 1980) and in the vadose zone (unpublished data) showed the NO_3 contamination in these three counties is predominantly from a nonpoint source and that the primary source is N fertilizer applied to cropland. Excessive N fertilization and irrigation exacerbate the contamination (Schepers et al., 1991). Nonpoint NO_3 contamination also has been identified in northern Holt County, in eastern Hall County, at Sidney in Cheyenne County, and near Oshkosh in Garden County (Fig. 4). The sources of the contamination in these areas also are agronomic and include applied commercial fertilizer (Exner and Spalding, 1979; Exner, 1990), injected sewage sludge (Spalding et al., 1993), and applied manure (Bryda, 1988).

Between 1985 and 1989 the Nebraska Department of Health conducted a study statistically designed to estimate the population at risk of ingesting NO_3-contaminated water from rural domestic wells. Nitrate concentrations in 2195 rural domestic wells from Nebraska's 93 counties averaged 6.6 mg/L NO_3-N and exceeded the MCL in 17.4% of the sampled wells (Spalding, 1991). This incidence of contamination was very similar to the ≈21% obtained by Exner and Spalding (1990) in their compilation of nonrandom data. The weighted mean exposure level for the rural population using private drinking water wells was 7.5 mg/L NO_3-N, whereas the weighted frequency of exposure to NO_3 levels exceeding the MCL was 20.4%.

As in the Ohio study (Baker et al., 1989), the level of NO_3 in the well was not associated with the distance between the well and septic system (Spalding, 1991). In

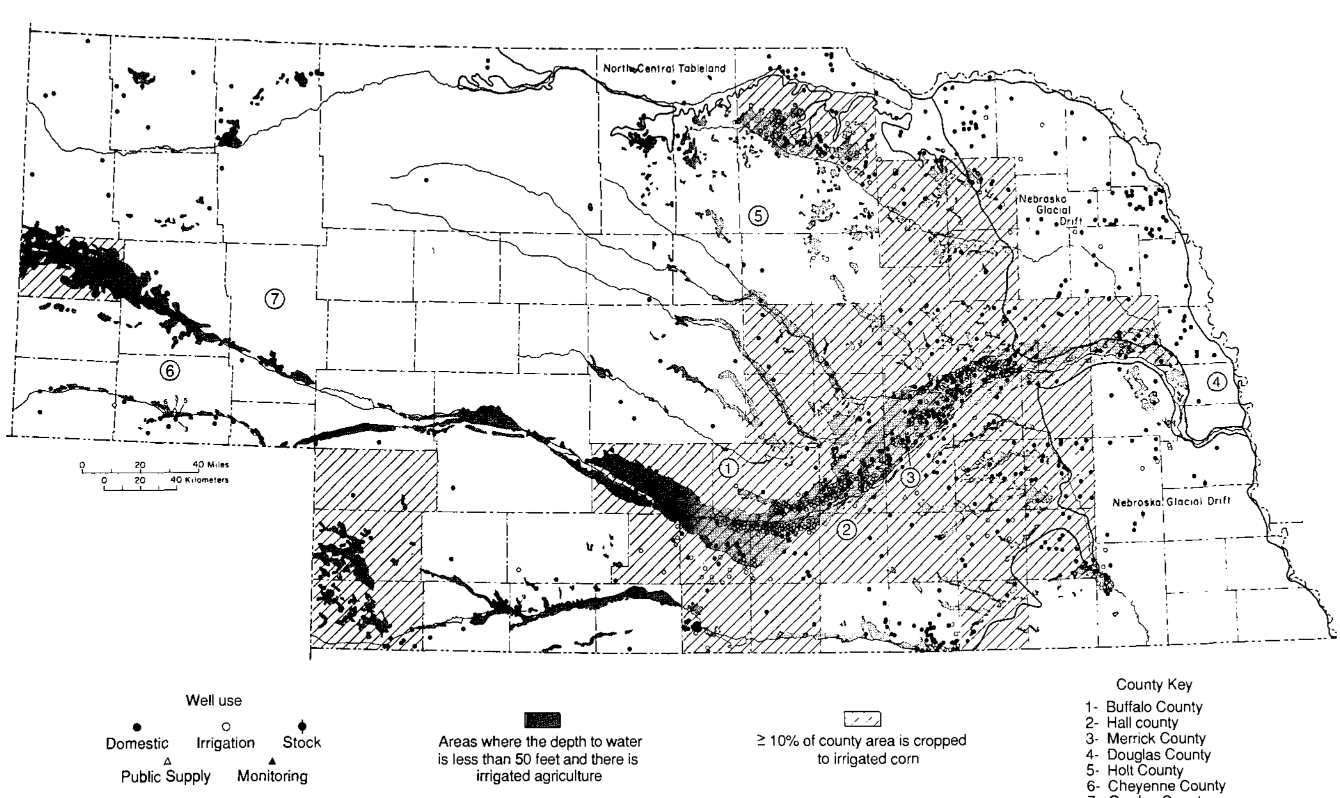

Fig. 4. Distribution of NO_3-N concentrations greater than 10 mg/L in Nebraska's groundwater (Exner and Spalding, 1990).

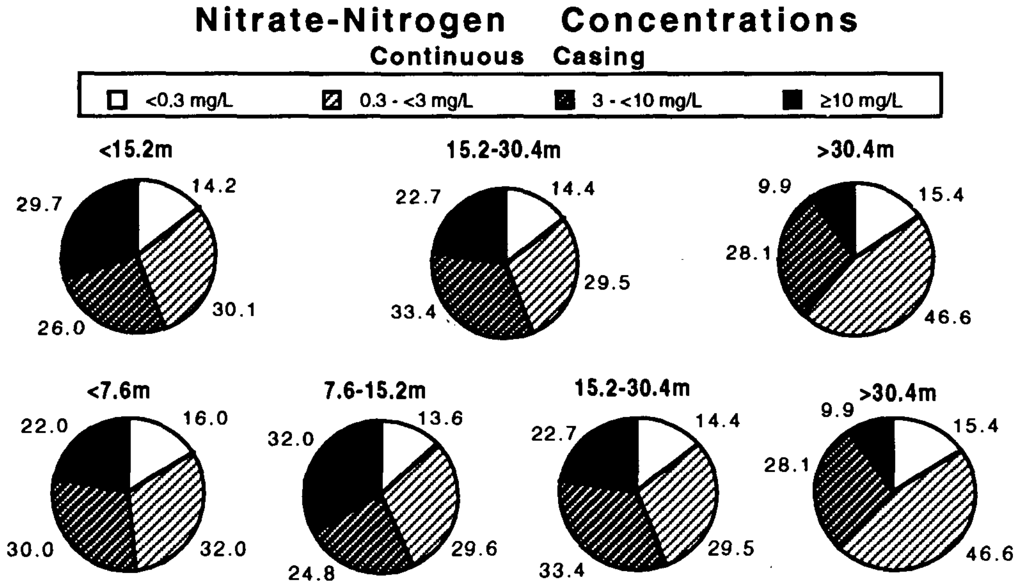

Fig. 5. Relationship between NO_3-N concentration and depth of continuously cased wells in Nebraska (Spalding, 1991).

general, these results support the belief that in rural areas septic systems are minor contributors of NO_3 relative to other sources (Hallberg, 1989). Clearly, septic systems can degrade groundwater quality (Robertson et al., 1991); however, it is somewhat unlikely that the limited radial influence of a domestic well will intercept the narrow transverse plume emanating from a septic system. In studies in densely populated, unsewered areas of Wisconsin, Tinker (1991) reported it was very difficult to intercept plumes from septic systems. Unless the well is situated close to a septic system or barnyard, or is immediately downgradient of these potential sources, it is unlikely that these sources will affect the quality of the well water.

Regression analysis of large data bases (Steichen et al., 1988; Baker et al., 1989; Kross et al., 1990; Spalding, 1991; Jennings, 1992, unpublished data) showed significant trends of decreased NO_3 concentrations with increased well depth. In Iowa, Hallberg (1989) attributed the vertical distribution of the NO_3 contamination, which appears primarily in the upper 15 m of the aquifer, to the hydrogeology of the vadose zone, to aquifer hydraulics, and to increased denitrification with depth in low dissolved oxygen bedrock aquifers. However, when shallow, continuously case wells (<15.2 m deep) in the Nebraska study (Spalding, 1991) were subdivided into wells <7.6 m deep and wells 7.6 to 15.2 m deep, the shallower wells had a lower frequency of contamination (Fig. 5). It is inferred that wells <7.6 m deep are located in areas with very shallow water tables. In a shallow water table area between the Platte and Elkhorn Rivers in Nebraska (4 in Fig. 4), where the groundwater is in seasonal contact with the fertilized soils, the incidence of NO_3 contamination is very low (Spalding, 1990). Gillham and Cherry (1977–1978) and Gillham (1991) have confirmed anoxic conditions in shallow groundwater, and Trudell et al. (1986) have shown that these conditions promote denitrification. Thus, groundwater denitrification is the suggested mechanism for NO_3 loss in both very shallow groundwater and in some deep groundwater.

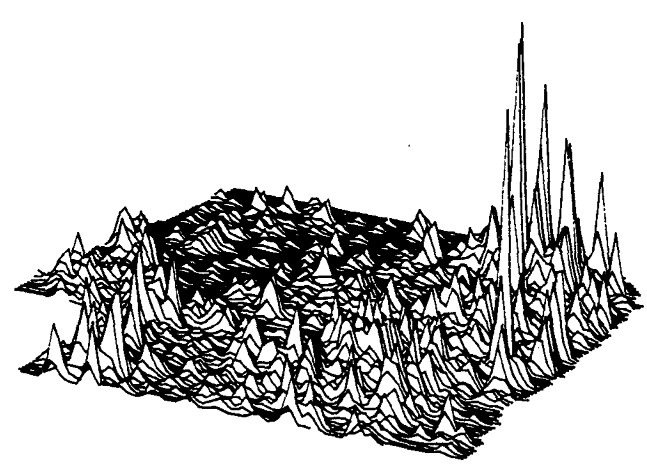

Fig. 6. Spline diagram of concentrations >10 mg/L NO_3-N in Nebraska groundwater.

Analysis of the Nebraska data (Spalding, 1991) also revealed distinct regional pattterns of NO_3 contamination (Fig. 6). Inordinately high NO_3 concentrations were prevalent in the rural domestic wells in the glaciated northeast—the same area that had the highest frequency of poor well construction. The majority of these poorly constructed wells are dug and lined with cement blocks, bricks, tile, or other open-jointed materials, which cause them to be vulnerable to surface contamination (Whitsell and Hutchinson, 1973). Both the incidence of contamination and the average NO_3 concentrations were higher in the wells with open-jointed casing. Nitrate concentrations in 60% of the 87 open-jointed wells exceeded the MCL, whereas concentrations in only 18% of the 221 continuously cased wells exceeded the MCL. The mean concentration of 25.4 mg/L NO_3-N in the wells with open-jointed casing was approximately four times higher than in the continuously cased wells. The incidence of total coliform contamination was three times higher in the wells with open-jointed casing and there was significant correlation ($r = +0.46$) between NO_3 concentrations and total coliform levels. This area of northeast Nebraska is adjacent and directly west of the Iowa area, which had the highest incidence of both poor well construction and NO_3 contamination in that state (Kross et al., 1990). In northwest Iowa it is difficult to obtain adequate groundwater; consequently, large diameter, open-jointed wells have been constructed to collect and store perched vadose zone water. In northeast Nebraska, however, the wells generally intersect the water table of the regional groundwater reservoir. The large diameter, open-jointed wells are typical of well construction in areas with fine-textured vadose zone sediments derived from loess where caving was not a problem.

The incidence of NO_3 contamination was very high in wells with open-jointed construction regardless of the age of the well (Fig. 7). This association also has been frequently reported (Steichen et al., 1988; Baker et al., 1989; Kross et al., 1990). Regardless of the water tightness of the casing, NO_3 as well as total coliform contamination were encountered slightly less often in newer (<20 yr old) wells than in wells more than 40 yr old. Similar associations between well construction, depth, age, and siting and the incidence of NO_3 contamination in studies in Iowa (Kross et al., 1990) and Ohio (Baker et al., 1989) support the widely held belief that modern well construction practices provide an effective barrier to surface contamination and can reduce the incidence of NO_3 contamination in domestic rural wells.

A highly suspected source of the NO_3 contamination in the wells in glaciated northeastern Nebraska is manure leachate. Leachate from the dissolution of manure in barnyards and corrals was the source of most of the NO_3 contamination in domestic and stock wells in glaciated southeastern Nebraska (Exner et al., 1985). The potential for development of cracks is much higher in the smectite-containing loess soils and unsaturated zone in glaciated eastern Nebraska than it is to the west. Nitrate and bacteria from the manure probably are transported in the unsaturated zone by preferential flow in desiccation cracks that develop in the fine-textured soils surrounding the well. These cracks can be more than 3 m deep and extend radially more than 20 m. The fine-textured soils and unsaturated zone of glaciated eastern Nebraska appear to form a generalized line of demarcation between this point-source area with wells containing very high NO_3 concentrations and nonpoint source areas to the west.

CONCLUSIONS

The skewed occurrence of large areas of NO_3-contaminated groundwater in the USA indicates that the groundwater in many intensely agricultural regions is not particularly vulnerable to NO_3 contamination. This distribution pattern is a consequence of both natural and man-induced factors. In the southeastern USA, abundant rainfall, high temperatures, and riparian soils with high organic C cause relatively rapid uptake of NO_3 and/or denitrification. Denitrification may also occur in the rich soils and subsoils of the Corn Belt but tile drainage appears more important in intercepting the downward movement of NO_3. The incidence of NO_3 contamination tends to increase in areas west of central Iowa and NO_3 concentrations are high in many sections of the Great Plains. Groundwater beneath irrigated, row-cropped areas with well-drained soils and permeable vadose zones in Minnesota, Washington, Arizona, California, and Nebraska is most impacted by dispersed-source NO_3 contamination.

A recurring theme throughout this review was that substandard well construction and improper siting of wells were associated with anomalously high NO_3 levels. Many of these wells were old dug wells. Samples from these wells are not representative of aquifer conditions and should not be used to assess dispersed-source contamination. These wells are highly vulnerable to surficial NO_3 contamination, because cracks in the fine-textured loess soils radiate from them during dry periods and provide preferential flow paths for contaminant transport to open joints in their casings.

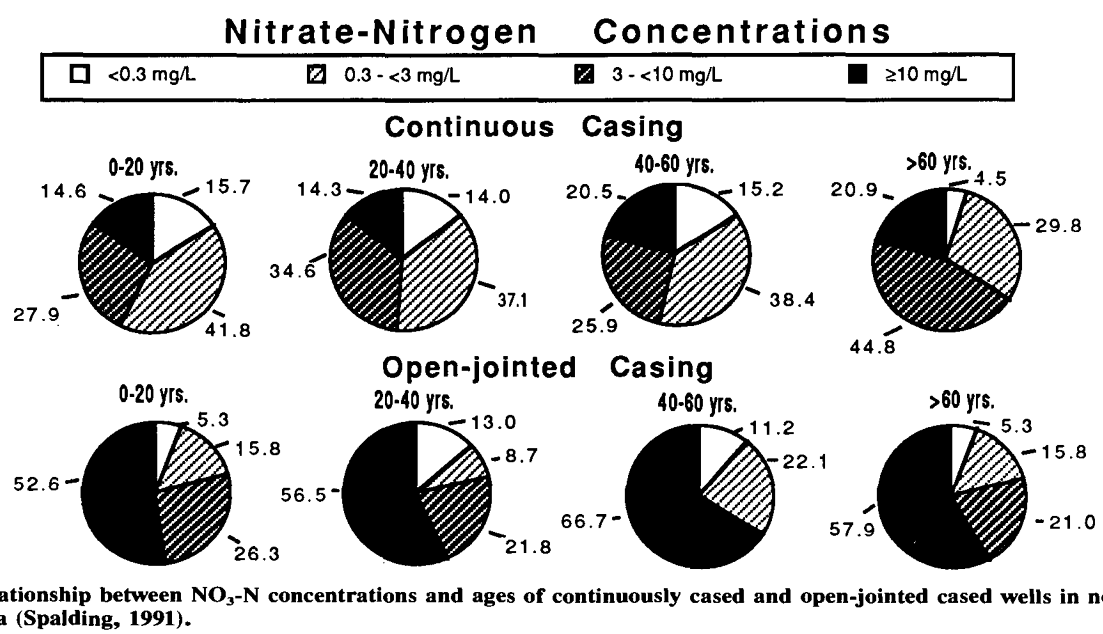

Fig. 7. Relationship between NO_3-N concentrations and ages of continuously cased and open-jointed cased wells in northeastern Nebraska (Spalding, 1991).

Although agrichemical contamination of groundwater is a growing concern both nationally and internationally, it must be kept in perspective. In most states the major areas of dispersed source, NO_3-contaminated groundwater have been delineated and are small in proportion to the total area of the state. As an example the estimated 300 000 ha underlain by nonpoint NO_3-contaminated groundwater in Nebraska comprise only 1.5% of the state's area. Although elevated NO_3 concentrations in wells in other areas of Nebraska could result from agrichemical contamination, there certainly is a strong case for point-source contamination in the poorly constructed and poorly sited wells in eastern Nebraska and the adjoining area of western Iowa.

The distribution of NO_3 in groundwater is controlled by a number of factors. They include source availability, thickness and composition of the vadose zone, precipitation, irrigation, vertical flow, aquifer heterogeneity, dissolved oxygen concentrations and electron donor availability, dispersion, and saturated thickness. Research must focus on the dynamics of groundwater NO_3. It is suggested that future assessments of the relative importance of vegetative uptake, denitrification, and geohydrology in limiting NO_3 contamination in deep and very shallow groundwater systems include measurements of dissolved oxygen, dissolved organic C, Fe, stable N isotopes, and microbial activity in discrete vertical intervals. Mariotti et al. (1988), Smith et al. (1991), and Spalding et al. (1993) are examples of such assessments.

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