Spatio-temporal Variability of Groundwater Nitrate Concentration in Texas: 1960 to 2010

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Nitrate (NO_3) is a major contaminant and threat to groundwater quality in Texas. High-NO_3 groundwater used for irrigation and domestic purposes has serious environmental and health implications. The objective of this study was to evaluate spatio-temporal trends in groundwater NO_3 concentrations in Texas on a county basis from 1960 to 2010 with special emphasis on the Texas Rolling Plains (TRP) using the Texas Water Development Board's groundwater quality database. Results indicated that groundwater NO_3 concentrations have significantly increased in several counties since the 1960s. In 25 counties, >30% of the observations exceeded the maximum contamination level (MCL) for NO_3 (44 mg L^{-1} NO_3) in the 2000s as compared with eight counties in the 1960s. In Haskell and Knox Counties of the TRP, all observations exceeded the NO_3 MCL in the 2000s. A distinct spatial clustering of high-NO_3 counties has become increasingly apparent with time in the TRP, as indicated by different spatial indices. County median NO_3 concentrations in the TRP region were positively correlated with county-based area estimates of crop lands, fertilized croplands, and irrigated croplands, suggesting a negative impact of agricultural practices on groundwater NO_3 concentrations. The highly transmissive geologic and soil media in the TRP have likely facilitated NO_3 movement and groundwater contamination in this region. A major hindrance in evaluating groundwater NO_3 concentrations was the lack of adequate recent observations. Overall, the results indicated a substantial deterioration of groundwater quality by NO_3 across the state due to agricultural activities, emphasizing the need for a more frequent and spatially intensive groundwater sampling.

Groundwater is a major drinking and irrigation water source in the United States (USGS, 2005). In 2005, about 68% of the nation's total groundwater development was used for irrigation (Hutson et al., 2004; USGS, 2005). Recent assessments have revealed a substantial deterioration of groundwater quality by NO_3 across the nation (Gurdak and Qi, 2006; Bronson et al., 2009; Showers et al., 2008; Dubrovsky et al., 2010; Welch et al., 2011). Irrigating with high-NO_3 groundwater and not accounting for NO_3 in irrigation water while deciding fertilizer application rates can lead to significant source water contamination. Ingestion of high-NO_3 groundwater can cause methemoglobinemia in infants less than 6 mo of age (Johnson and Kross, 1990). Based on the human health risks, the NO_3 maximum contamination level (MCL) is established at 44 mg L^{-1} NO_3 (equivalent to 10 mg L^{-1} NO_3–N) (USEPA, 2006). Growing concern over human health risks from groundwater NO_3 contamination has propelled numerous initiatives to understand NO_3 dynamics in diverse hydrologic regimes.

Soil NO_3 originates from various natural (soil N and atmospheric deposition) as well as anthropogenic (mostly a mixture of NO_3 and ammonium fertilizers) sources (Wakida and Lerner, 2005). Due to its solubility and mobility, NO_3 can easily leach to groundwater and persist for decades depending on the hydrologic regime (Nolan et al., 2002). A variety of factors influence NO_3 entry to groundwater, including climate, land use, aquifer characteristics, and groundwater table depth and recharge patterns (Masetti et al., 2008). A critical review of potential factors that affect fate and transport of NO_3 in soils is therefore essential to address groundwater NO_3 contamination issues and to formulate ameliorative strategies.

According to Dubrovsky et al. (2010), groundwater NO_3 concentration above 4.4 mg L^{-1}, a concentration deemed as national background for NO_3, indicates anthropogenic influences. Nitrogenous fertilizers and manures used in agriculture are identified as a major source of NO_3 to groundwater (Almasri and Kaluarachchi, 2007; Ghiglieri et al., 2009; Howden et al., 2011a,b; Mencio et al., 2011). Babiker et al. (2004) found that high NO_3 levels (>44 mg L^{-1}) were mostly associated with vegetable fields in Japan. In Iowa, Steinheimer et al. (1998) found significantly high

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Abbreviations: GMA, groundwater management area; LISA, local indicators of spatial association; MCL, maximum contamination level; NLCD, National Land Cover Dataset; SHP, Southern High Plains; TRP, Texas Rolling Plains; TWDB, Texas Water Development Board.

groundwater NO_3 concentrations from agricultural lands. The effect of irrigation practices on NO_3 contamination is well documented (Novakova and Nagel, 2009). According to Costa et al. (2002), well drained soils with high irrigation and fertilizer input have a higher risk of inducing NO_3 contamination.

Nitrate is a ubiquitous groundwater contaminant in Texas (Nolan et al., 2002), with high concentrations mostly reported from different counties of the Southern High Plains and the Rolling Plains (Hudak, 2000; Scanlon et al., 2003a,b, 2008; Bronson et al., 2009). Hillin and Hudak (2003) analyzed groundwater NO_3 concentration data in the Texas Rolling Planes (TRP) for 12 years between 1936 and 1997 and found that in each year at least 37% of observations exceeded the NO_3 MCL. The major source of NO_3 in Texas groundwaters is unclear. A variety of natural and anthropogenic sources have been suggested on the basis of the stable N isotope (^{15}N) natural abundance method (Singleton et al., 2005; Scanlon et al., 2008). In a study conducted in Runnels County in Texas, Kreitler and Jones (1975) concluded that about 66% of the NO_3 in groundwater had natural sources. In general, the $\delta^{15}N$ values of N from commercial fertilizers, soil, precipitation, and animal wastes range from −7 to +8‰, +5 to +7‰, 12 to +3‰, and greater than +10‰, respectively (Kellman and Hillaire-Marcel, 2003; Townsend et al., 2003). However, considerable overlap in $\delta^{15}N$ between N sources makes their accurate identification difficult using N isotopes. Additionally, due to inherent soil spatial variability and variability of microbial transformations, it is difficult to pinpoint NO_3 sources (Roadcap et al., 2002).

Shallow groundwater NO_3 contamination has been identified by numerous studies in the United States and around the world (Rekha et al., 2011; Welch et al., 2011; Grimaldi et al., 2012). Hudak (2000), Scanlon et al. (2003b), Gurdak and Qi (2006) and Saphr et al. (2010) found higher groundwater NO_3 concentrations in shallow groundwater wells, an observation that led them to conclude a surface origin of NO_3, possibly agricultural, in Texas. Hudak's (2000) study identified a regional pattern in groundwater NO_3 concentrations in Texas. However, the study focused on only one decade (1990–1998), and, although a spatial trend was identified, no rigorous spatial statistical methods were used for quantitative assessment of spatio-temporal trends. Hillin and Hudak (2003) found that higher median NO_3 concentrations, exceeding the MCL, in the Seymour aquifer were mainly associated with agricultural croplands. According to Kreitler (1975), high groundwater NO_3 concentrations in the TRP region, before the extensive fertilizer application that began in the 1960s in Texas, ensued mainly from soil N transformation processes. Bartolino (1994) attributed high NO_3 concentrations to symbiotic N fixation by leguminous plants and subsequent flushing of NO_3 into groundwaters due to increased recharge. Conversion of native rangeland and grasslands to crop lands has also been suggested as a potential cause of elevated groundwater NO_3 concentrations in the TRP (Price, 1979; Ewing et al., 2004).

The goal of this study was to characterize long-term (1960–2010) spatio-temporal trends in groundwater NO_3 contamination across Texas counties, with special emphasis on 22 counties in the TRP region, by compiling NO_3 concentration data from the Texas Water Development Board's (TWDB) groundwater quality database. The specific objectives of this study were (i) to identify current data gaps and future data needs to assess spatio-temporal trends in groundwater NO_3 concentration, (ii) to analyze spatial trends in groundwater NO_3 concentrations between 1960 and 2010, (iii) to delineate high NO_3 risk areas, and (iv) to identify the potential factors that influenced groundwater NO_3 contamination. Results presented herein report spatio-temporal distribution of groundwater NO_3 concentrations in Texas counties without referring to aquifers.

Materials and Methods

Study Area

Texas is characterized by a diverse biogeography and climatic conditions. Climatic conditions in Texas, due to its large size, vary widely across the state, with mostly arid/semiarid conditions in the west to humid, subtropical conditions in the east. There are nine major and two minor aquifers in Texas, underlying about 76% of the land area and supplying about 60% of the total water used in the state (Texas Groundwater Protection Committee, 2003; Texas Water Development Board, 2007). About 79% of the total extracted groundwater in Texas is used for agricultural irrigation (Fig. 1a) (USGS, 2005). About 99% of the rural households in the state rely on groundwater for their drinking water supply (Texas Water Development Board, 2007). Agriculture is a major industry in Texas, which ranks second in the United States in total agricultural production (6.8% of the total US production) after California (USDA, 2010).

The TRP region in north-central Texas is comprised of 22 counties (Fig. 1a) (Ashworth and Hopkins, 1995). Land cover in the TRP is dominated by shrub, grass, and agricultural croplands. Primary lithotype included unconsolidated Quaternary alluviums comprised of fine- to coarse-grained gravel, fine- to coarse-grained sand, silt, clay, and caliche (Harden and Associates, 1978). The majority of groundwater in the TRP region is used for domestic, public use, and irrigation purposes (Fig. 1a). Most of the groundwater wells are used for water withdrawal (Fig. 1b). Principal land cover types in this region include shrubs and crop lands (Fig. 1c). Average well yields in this region vary around 1.13 $m^3 min^{-1}$ (Texas Water Development Board, 2003).

Groundwater Database

The TWDB database contains a large amount of groundwater quality data, including concentrations of NO_3 and several other solutes dating back to 1896. The groundwater database contains information about 33 major and minor chemical species and several water quality parameters for Texas aquifers. The TWDB maintains the groundwater database for the state as a part of the Water Information Integration and Dissemination system with information about over 120,000 water wells, springs, and oil and gas test sites in Texas (Fig. 1a). Groundwater quality sampling protocols are described in detail elsewhere (Nordstrom and Beynon, 1991; Hopkins, 1995; Boghici, 2003). Briefly, before sampling, wells are purged until pH, specific conductance, and temperature readings stabilized. Groundwater samples were collected near the wellhead to ensure representative samples. Wells equipped with faucets at the wellhead were preferred for sample collection. Water samples were filtered using a pressure tank system before analysis and preserved with acid for analysis of cationic species. Geochemical characterizations are performed by automated colorimetric and/or chromatographic methods (Hudak, 2000).

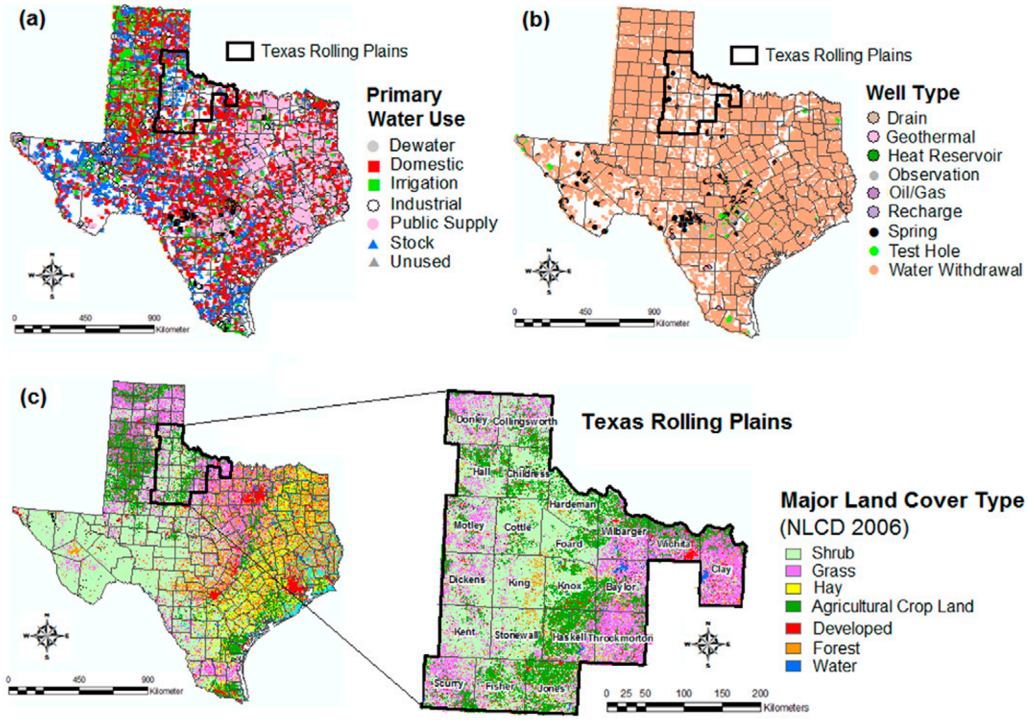

Fig. 1. (a) Primary water use and (b) well type of the groundwater wells corresponding to groundwater NO_3 observations that have been included in the study and (c) major land cover classes in Texas. (Source: Texas Water Development Board, National Land Cover Dataset, 2006).

Statistical Analyses

Groundwater NO_3 concentration information between 1960 and 2010 was obtained from the TWDB's groundwater quality database. Due to a lack of adequate per-county NO_3 data for any single year, NO_3 data were aggregated into 10-yr periods (1960s, 1970s, 1980s, 1990s, and 2000s) and compiled in ArcView v.9.3 (Table 1). Before analyses, quality control assessments were performed in two stages. Each groundwater quality observation in the TWDB database is assigned a code indicating if sampling and geochemical analyses were performed following established protocols. Data were screened according to reliability codes, and only reliable observations were selected for further examination. After data screening, each groundwater NO_3 observation was subjected to ion charge balance calculations, and only the observations with less than ±5% charge balance error were included in the analysis. For groundwater observation wells that had multiple observations in a decade, the most recent observation that passed quality control assessments mentioned above was selected for further statistical and spatial analysis. Inferences drawn about spatio-temporal patterns of groundwater NO_3 concentration in this study are based on NO_3 concentration observations representing corresponding groundwater well locations on the maps. County median NO_3 concentrations, or any other descriptive statistic for NO_3, were calculated by summarizing the all groundwater NO_3 observations separately for each county. Temporal changes in groundwater NO_3 concentrations were assessed by computing county-based descriptive summaries for each decade with the Statistical Analysis System. Due to non-normality of data, intra- and interdecadal comparisons between the counties were performed by the Wilcoxon Mann-Whitney test at the 95% significance level. Differences in median concentrations were tested by the Hodges-Lehmann estimator.

Table 1. Descriptive summary of decadal groundwater nitrate concentration in Texas and the Texas Rolling Plains.

Time period	Sample size	Minimum	25th Percentile	Median	75th Percentile	Maximum	SD
		mg L^{-1} as NO_3					
Texas							
2000s	6,227	0.0	0.1	3.4	11.0	614.0	27.3
1990s	8,790	0.0	0.2	2.5	10.6	1483.7	33.5
1980s	10,185	0.0	0.0	1.4	8.9	1561.8	33.1
1970s	15,544	0.0	0.4	1.4	13.0	2162.0	58.1
1960s	20,342	0.0	0.4	1.5	9.0	2640.0	70.8
Texas Rolling Plains							
2000s	213	0.0	9.3	30.6	62.4	197.4	40.8
1990s	422	0.0	5.1	27.7	58.3	1483.7	82.1
1980s	565	0.0	0.0	22.6	36.2	587.9	54.3
1970s	1,775	0.0	21.0	23.0	65.0	1480.0	63.0
1960s	2,395	0.0	0.4	13.0	43.0	1490.0	92.9

The groundwater NO_3 concentrations were also assessed by the 16 groundwater management areas (GMAs) of the state. The extent of groundwater NO_3 contamination in a county was assessed by computing the county-based percentages of groundwater NO_3 observations exceeding the NO_3 MCL for each decade. Percentages were calculated by dividing the number of observations exceeding the MCL by the total number of observations in that county and presented as spatial maps for each decade. To identify statewide high NO_3 risk zones and to test for the existence of spatial autocorrelation between counties, two spatial indices, Moran's I and Local Indicators of Spatial Association (LISA), were computed using the county-based percentages of observations exceeding the MCL as described above. For both indices, a first-order queen contiguity between the adjoining counties was assumed to derive the spatial weight matrix. Moran's I is an extension of Pearson's product–moment correlation coefficient; its numerator represents a covariance function, and the denominator represents the sample variance (Moran, 1950). Moran's I was computed using following the equation (Moran, 1950):

$$\mathrm{I} = \frac{n}{S_o} \frac{\sum_{i=1}^{n}\sum_{j=0}^{n} \mathrm{W}_{ij}(\mathrm{x}_i - X)(\mathrm{x}_j - X)}{\sum_{i=1}^{n}(\mathrm{x}_i - X)^2} \quad [1]$$

in which

$$\mathrm{S}_o = \sum_{i=1}^{n}\sum_{j=1}^{n} \mathbf{W}_{ij} \quad [2]$$

where n is the total number of observations, i and j are locations, $\mathbf{W}_{ij}$ is the spatial weight matrix, S_o is the product sum of the spatial weight matrix, x_i is the value at location i, x_j is the value at location j, and X is the mean of value x.

The spatial matrix ($\mathbf{W}_{ij}$) is preferably row-standardized and by convention equals zero. Moran's I varies from +1 (strong positive spatial autocorrelation) to −1 (strong negative spatial autocorrelation) with an expected value of 0 under the null hypothesis, suggesting a lack of spatial autocorrelation (random pattern) (Ping et al., 2004). However, Moran's I is a global indicator that yields a single statistic for the entire area by "averaging out" local variations, which in large data sets give rise to spatial instability or local nonstationarity phenomena (Anselin, 1995). In this regard, LISA provides a better indication of clustering of the spatial units by calculating local Moran's I for each unit. This involves decomposition of the global statistic (Moran's I) into its constituent parts (local indices), whose sum is proportional to the global statistic. The LISA is a distance-based method that quantifies the extent of clustering of the "high" or "low" value of the parameter of interest and how the values in the neighboring locations vary within a specified bandwidth (distance). Four scenarios may emerge under LISA: (i) "high-high," where high-NO_3 counties are associated with high-NO_3 counties; (ii) "low-low," where low-NO_3 counties are associated with low-NO_3 counties; (iii) spatial outliers, where high-NO_3 counties are associated with low-NO_3 counties and vice versa; and (iv) locations with no significant spatial clustering. The significance in spatial association is derived through conditional randomization in an iterative manner (Hubert et al., 1985).

To understand the influence of agricultural practices on groundwater NO_3 concentrations, statewide land cover information was obtained from the National Land Cover Dataset (NLCD) for 2006 (Fry et al., 2011). County-based area estimates of fertilized crop lands and irrigated lands for each TRP county were obtained from the USDA National Agricultural Statistics Service (NASS, 2007). Due to non-normality of NO_3 concentration data, Spearman rank correlations were performed between the county-based median NO_3 levels and area estimates of NLCD crop lands, fertilized crop lands, and irrigated lands to understand the influence of agricultural practices on groundwater nitrate levels. The latter two were used as surrogates to the actual amounts of fertilizer input and irrigation water.

Results and Discussion

Spatio-temporal intensity of groundwater quality monitoring by the TWDB has recently decreased, leading to a significant reduction in available water quality information. For example, only about 4% of the groundwater observation wells that were monitored in the 2000s have representations in other decades. In the 2000s, no observations were available for seven counties (Archer, Palo Pinto, Rockwall, Shakelford, Stephens, Throckmorton, and Young), whereas only Rockwall County was without an observation in the 1990s.

Some of the wells that recorded high NO_3 concentrations in one decade had not been monitored in any other decade. For example, two wells that recorded high NO_3 concentrations of 195 mg L^{-1} (Baylor County) and 1483 mg L^{-1} (Jones County) in the 2000s and the 1990s, respectively (both located in the TRP), did not have records from the earlier decades. On the other hand, no recent observations are available for wells located in the Brown and Runnels Counties where the highest NO_3 concentrations (>2000 mg L^{-1}) were recorded in the 1960s and the 1970s.

In the TRP region, NO_3 concentration data were available for about 2400 and 1800 wells in the 1960s and the 1970s, respectively, as compared with 422 and 213 wells in the 1990s and the 2000s, respectively. Similarly, in the 1970s, about 440 and 480 groundwater wells were monitored for water quality in Haskell and Knox Counties (which recorded the highest NO_3 concentrations in the state), respectively, as compared with only 19 and 60 wells from the respective counties in the 2000s, indicating a significant reduction in the intensity of NO_3 monitoring in recent years. This emphasizes the need for a more spatially intensive monitoring at more frequent intervals to understand the spatio-temporal changes in groundwater quality and to address related environmental issues. This will not only enhance awareness among general public but will also attract attention of the concerned authorities to identify NO_3 risk areas of the state and develop appropriate remedial measures.

The statewide median NO_3 concentration was significantly (p < 0.01) higher in the 2000s than in the previous decades, indicating a substantial deterioration of groundwater quality over time (Table 1). The median NO_3 concentrations in the TRP were much higher than the statewide medians in all decades of the study. A noticeable concentration of high NO_3 occurrences (at concentrations above or close to MCL) has

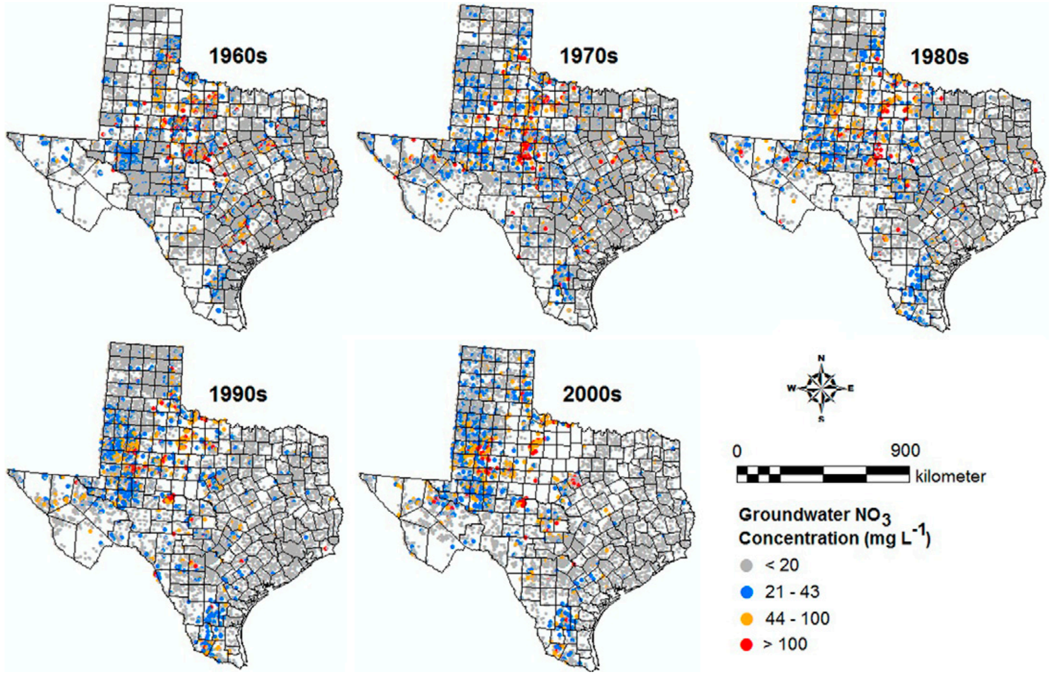

Fig. 2. Decadal groundwater NO_3 concentrations (mg L^{-1} as NO_3) in Texas. Each dot on the map indicates the most recent groundwater NO_3 observation representing the groundwater quality of the corresponding groundwater observation well at that location in that particular decade. White regions on the maps indicate areas without groundwater NO_3 observation for that decade; however, it does not imply the absence of any groundwater observation well in those regions. (Data source: Texas Water Development Board.)

become apparent in several west- and north-central counties in all decades since the 1960s (Fig. 2). The increase in groundwater NO_3 concentration since the 1960s probably stemmed from widespread fertilizer and organic waste (enriched in N) application to enhance agricultural production, which began in the mid-1960s (Bartolino, 1994).

In 2006, crop lands comprised about 11% of total land cover in Texas and were among the dominant land cover types in the north-central and west-central counties of the state. Twelve counties located in the Southern High Plains (SHP) had more than 50% of land area under agricultural land cover (Fig. 3a). In the 2000s, about 15% of the groundwater NO_3 observations obtained from the crop lands exceeded the MCL, as compared with only about 5% from other land cover types. In the TRP region, the highest median NO_3 concentrations in the 2000s were observed in Knox County (81 mg L^{-1} NO_3) and Haskell County (79 mg L^{-1} NO_3), both having high (>35) percentages of agricultural crop land (Fig. 3b). Out of all the observations made in the agricultural croplands in the TRP region in the 2000s, about 60% exceeded the NO_3 MCL, and a majority of them were from Knox and Haskell Counties. These findings are in agreement with Scanlon et al. (2003a), who suggested that NO_3 contamination in the TRP region resulted from agricultural activities. Significant correlations (Spearman $\rho = 0.26$; $p < 0.001$) between county-based median NO_3 concentration and percent crop lands in the 2000s indicated a positive correlation between agricultural practices and groundwater NO_3 concentrations. In general, high NO_3 concentrations (close to or above the MCL) have mostly been found in counties with >25% crop lands. High NO_3 concentrations (>30 mg L^{-1} NO_3) in southern Texas counties (Brooks, Duval, Hidalgo, Jim Hogg, Jim Wells, and Starr) probably resulted from NO_3 leaching from irrigation return flows from the agricultural lands in these counties (Fig. 3b). There are about 300 irrigation wells in this region draining through highly permeable geologic formations, leading to enhanced NO_3 transport to the groundwaters (Texas Water Commission, 1989).

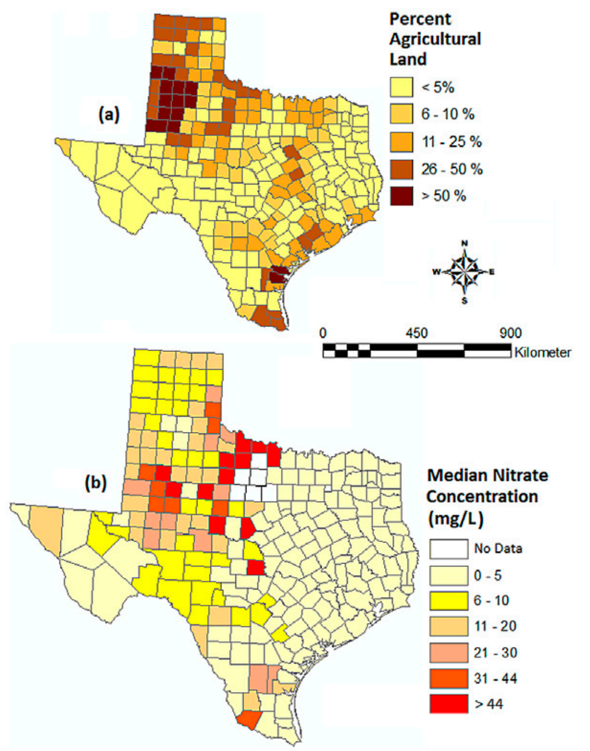

Fig. 3. Countywise (a) percentages of crop lands in 2006 and (b) county median NO_3 concentrations (mg L^{-1} NO_3) in the 2000s. For each county, percentage crop land was computed as the ratio of crop land area to the total land cover area in the county; county median NO_3 concentrations were computed by considering all of the most recent groundwater NO_3 observations for each county.

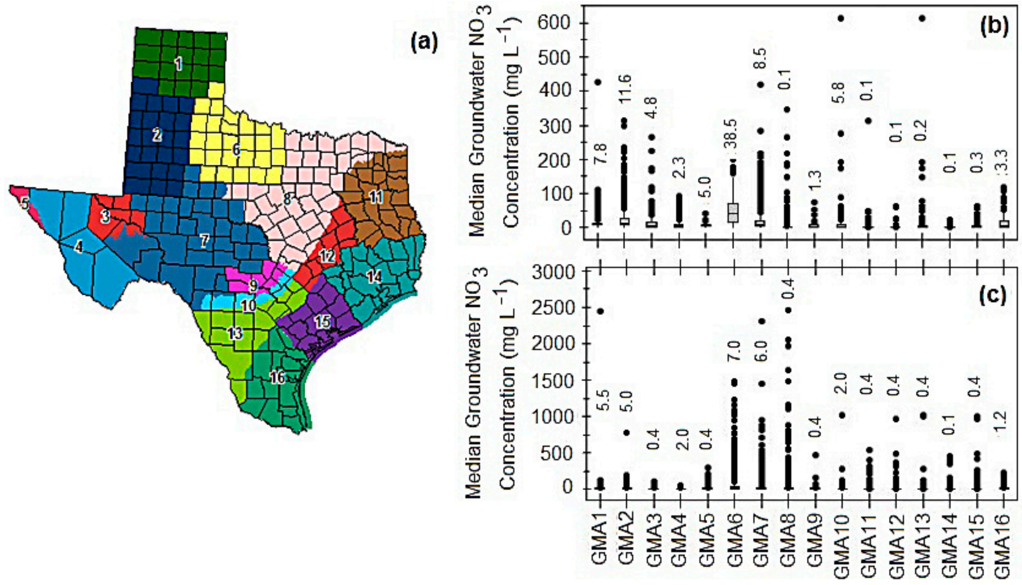

Fig. 4. (a) Distribution of 16 groundwater management areas (GMAs) in Texas and the GMA-wise variability in groundwater NO_3 concentrations (mg L^{-1} NO_3) for (b) 2000s and (c) 1960s (Data Source: Texas Water Development Board). (b and c) Numbers indicate median NO_3 concentrations; red dots indicate outliers. Median NO_3 concentration for each GMA was calculated from all NO_3 observations from all counties that constitute the GMA.

In eastern and central Texas, significantly low groundwater NO_3 concentrations (<10 mg L^{-1} NO_3) were observed in all decades. This is probably due to (i) the presence of forest ecosystems that do not receive fertilizers, (ii) N uptake by plants, and (iii) denitrification processes favored by the warm, moist conditions prevalent in these regions and high soil organic matter (Hudak, 2000). The variation in precipitation (ranging from about 120 cm in the east to <40 cm in the west of Texas) and evapotranspiration, which affects microbial N transformations, coupled with evaporative processes might have affected NO_3 concentrations in these regions. The presence of restrictive (impervious) soil layers associated with urbanized and developed areas in these regions might also have accounted for reduced infiltration and NO_3 leaching. However, the occurrence of some high NO_3 concentrations around the metropolitan areas in this region in the 1970s and the 1980s suggested NO_3 input from the lawn fertilizers, septic systems, and leaking municipal sewers.

Assessment of the extent of the groundwater NO_3 contamination by GMAs revealed significantly ($p < 0.01$) higher median NO_3 levels in all the GMAs in the 2000s (Fig. 4a–c). Consistently high median NO_3 concentrations found in GMA 6, comprising the TRP, indicated that groundwater NO_3 contamination has been a long-standing problem in the TRP counties. The high median NO_3 concentrations in GMAs 1 and 2 indicate agricultural sources of NO_3 in these regions. In GMAs 8, 11, 12, and 14, the median NO_3 concentrations were <1 mg L^{-1} NO_3, probably due to a lack of significant N inputs and/or denitrification processes.

The decadal pattern of the county-based percentages of observations exceeding the NO_3 MCL indicated that the number of counties with high groundwater NO_3 observations has noticeably increased over time (Fig. 5a and b). There were 25 and 20 counties with >30% observations exceeding the MCL in the 2000s and 1990s, respectively, as compared with only eight counties in the 1960s. Out of 25 counties that had >30% observations above MCL in the 2000s, 9 were located in the TRP, whereas only 5 such counties were found in the 1960s. The number of counties with >30% observations above MCL increased and the number of counties with 20 to 30% observations above MCL decreased over time (Fig. 5b). These observations indicated that, over time, the groundwater has been increasingly contaminated by NO_3 in the TRP region.

Groundwater NO_3 concentrations appeared spatially autocorrelated, as suggested by a strong positive Moran's I in all decades (Fig. 6). Furthermore, the increasing trend in Moran's I between the 1960s and the 1990s indicated that the spatial clustering phenomenon has become increasingly prominent over time. Moran's I is computed from a spatial contiguity matrix, which is constructed from the values of neighboring spatial units (counties). An absence of values in neighboring units yields a lower Moran's I. A sudden drop in Moran's I in the 2000s was probably caused by the absence of NO_3 concentration data from seven counties in the state. Moran's I has been extensively used in assessing spatial and temporal trends in water quality, quantity, and use (Franczyk and Change, 2009; Salendu, 2010; Shomar et al., 2010; Machiwal et al., 2012). Witheetrirong et al. (2011) used Moran's I to assess effect of soil texture and agricultural activities on spatial patterns of groundwater NO_3 contamination in Thailand. Sohel et al. (2010) used Moran's I to depict spatial autocorrelation patterns of groundwater arsenic concentrations in rural Bangladesh. In both studies, the Moran's I statistic appeared to be an appropriate measure to analyze spatial patterns in groundwater quality. Moran's I offers a "smoothed" global view without taking into account the local variations in spatial autocorrelation. In contrast, LISA provides "localized" spatial representation by identifying the exact locations of spatial data clusters and data outliers (i.e., the association of similar and dissimilar values) (Franczyk and Change, 2009). The LISA patterns across the state indicated a significant clustering of counties with high (>30) percentages of observations exceeding the MCL in the TRP and parts of SHP, a pattern more apparent in recent decades (Fig. 7). The decadal LISA patterns clearly identified the TRP and parts of the SHP as NO_3 "hotspots," indicating the need for a detailed scrutiny of the extent of NO_3 contamination in these regions and identify-

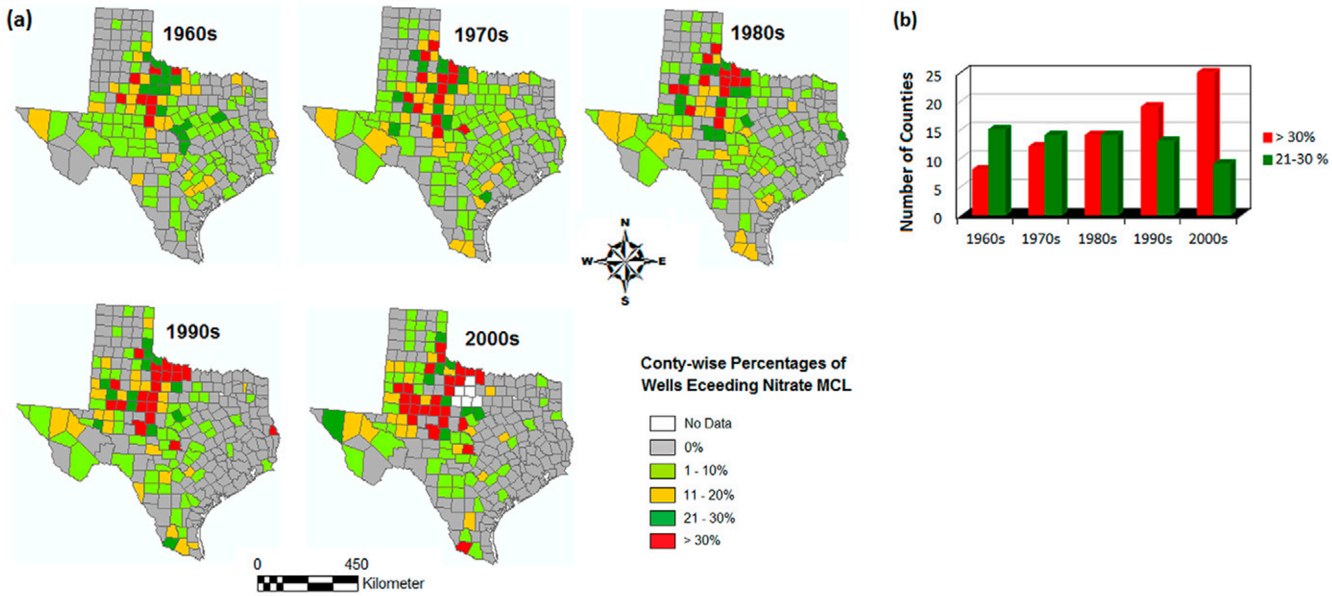

Fig. 5. (a) Decadal scenario of county-based percentages of groundwater NO_3 observations exceeding the maximum contaminant level (MCL) and (b) temporal pattern of number of counties with >30% and 20 to 30% NO_3 observations exceeding the MCL. The method for calculating county-based percentages is described in the Materials and Methods section. White regions in (a) indicate counties with no groundwater NO_3 observation in the respective decades, which, however, do not imply absence of groundwater wells in those regions. (Data source: Texas Water Development Board.)

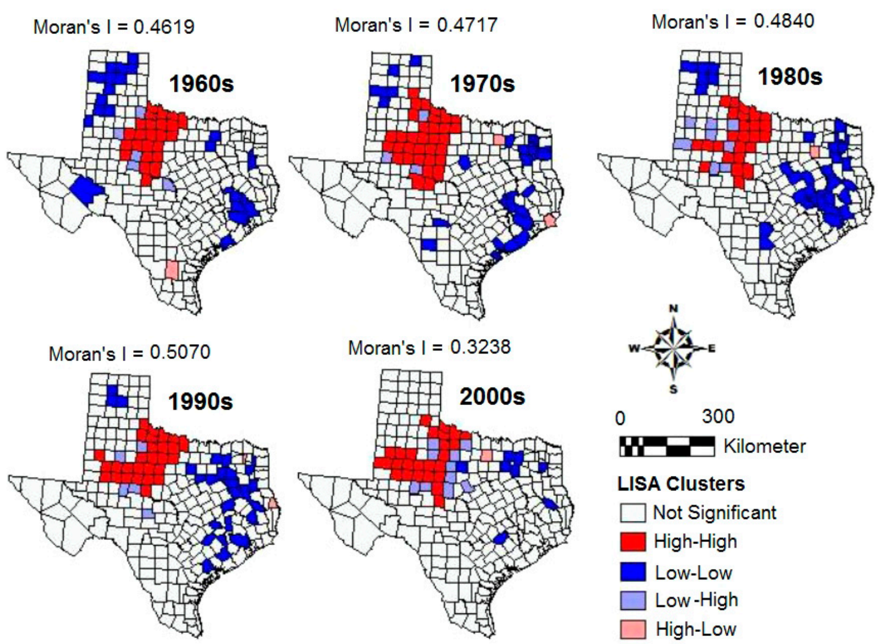

Fig. 6. Spatial cluster maps of county-based percentages of groundwater NO_3 observations exceeding NO_3 maximum contaminant level as indicated by Moran's I and LISA (Local Indicators of Spatial Autocorrelation) between the 1960s and the 2000s.

ing potential causes. The LISA maps also identified significant clustering of low-NO_3 counties in the eastern parts of the state, reiterating our earlier observations about low NO_3 occurrences in these regions.

The median NO_3 concentration in the TRP was significantly higher ($p < 0.05$) than the state median in the 2000s, indicating a different hydrochemical setting of the TRP as compared with the rest of the state (Table 1). In the TRP region, about 24% of NO_3 observations exceeded the MCL in the 1960s as compared with about 40% in the 2000s (Fig. 7). The majority of high NO_3 occurrences (>100 mg L^{-1} NO_3) in the 2000s were found in Knox, Haskell, Wilbarger, Wichita, Hardeman, Motley, and Collingsworth Counties. In Haskell and Knox Counties, all the observations in the 2000s exceeded the MCL, as compared with only about 21 and 24% for the respective counties in the 1960s. In Wilbarger, Wichita, and Baylor Counties, about 64, 60, and 50% of the observations, respectively, were above the MCL in the 2000s, as compared with about 25, 33, and 30%, respectively, in the 1990s. In Knox, Haskell, Wilbarger, and Wichita Counties, the median NO_3 concentrations exceeded the MCL in all decades, with the highest county medians found in Haskell and Knox Counties (Table 2).

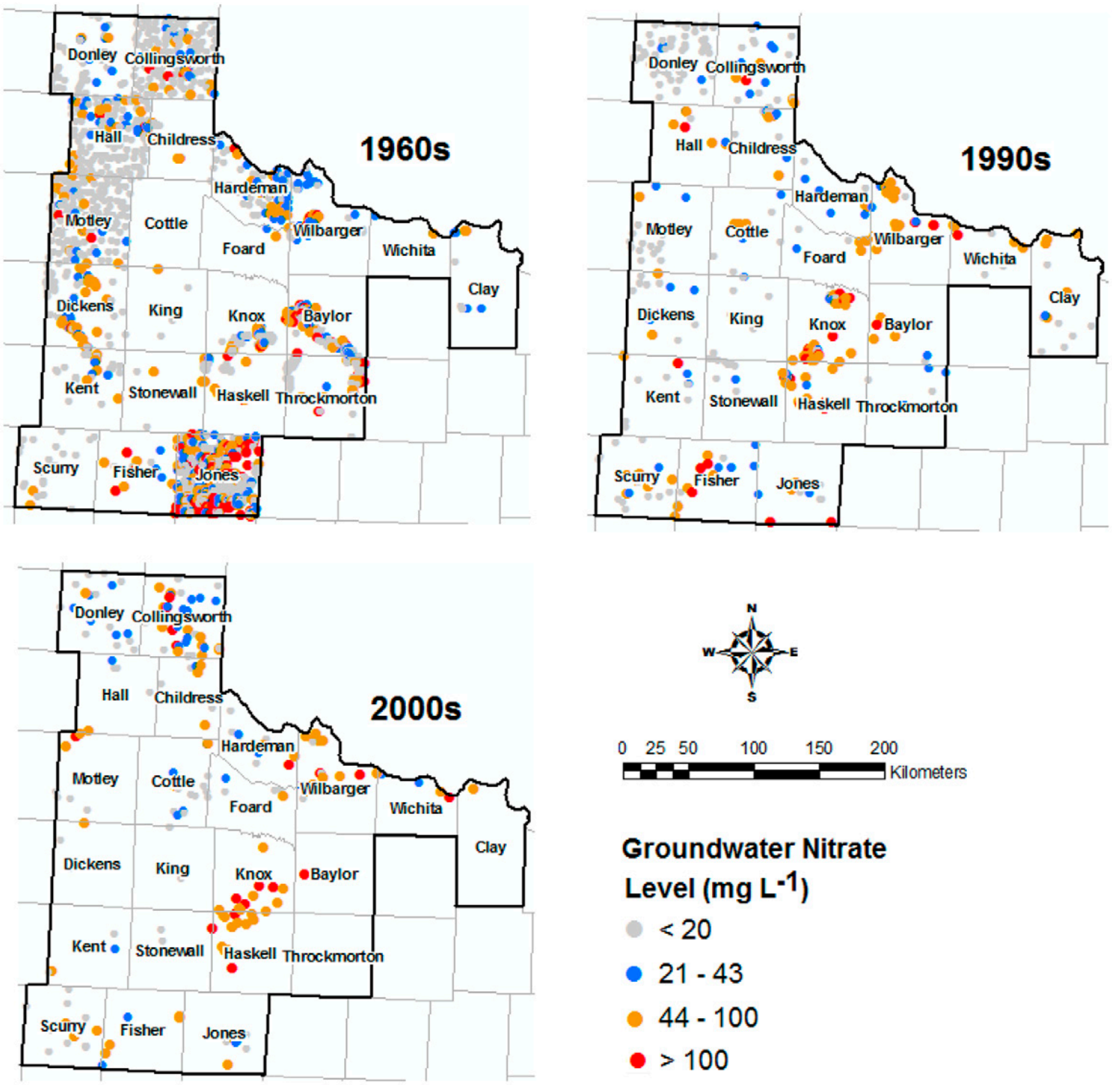

Fig. 7. Spatio-temporal distribution of groundwater NO_3 concentrations (mg L^{-1} NO_3) in the Texas Rolling Plains. (Data source: Texas Water Development Board.)

Between the late 1980s and the 2000s, the estimated area of fertilized croplands increased in 12 counties (Knox, Haskell, Wilbarger, Wichita, Jones, Hardeman, Hall, Foard, Fisher, Dickens, Cottle, and Collingsworth) in the TRP region (Fig. 8a). During the same period, the estimated area of irrigated land increased in eight counties (Wilbarger, Wichita, Haskell, Hall, Hardeman, Donley, Collingsworth, and Childress) (Fig. 8b). Spearman rank correlations performed between county median NO_3 concentrations and areas of (i) fertilized croplands, (ii) irrigated lands, and (iii) the NLCD crop lands were statistically significant (α = 0.05) for the TRP region, indicating a negative impact of agricultural activities on groundwater NO_3 concentrations (Table 3). Furthermore, an increasing trend in the correlation coefficient between fertilized croplands and NO_3 concentrations between late the 1980s and the mid-2000s indicated a stronger association between agricultural activities and groundwater NO_3 concentrations with time in the TRP region.

The amount of fertilizer applied to crop lands in Texas has increased over time (USDA, 1992, 1996, 2007). For example, N fertilizer input for cotton, a major agricultural crop in Texas, increased from about 99,000 tons in 1990 to 157,400 tons in 2007 (Fig. 9a), suggesting a high potential for NO_3 accumulation at or below the root zone. The amount of groundwater that has high NO_3 concentration pumped from the Seymour aquifer and used for irrigation purposes has also increased over time (Fig. 9b). In the TRP, the high-NO_3 groundwater counties (i.e., with median NO_3 levels exceeding the nitrate MCL), such as Knox, Haskell, and Wilbarger, have been associated with high groundwater irrigation since the mid-1980s (Fig. 9c). This indicated that, over time, increased N fertilizer input coupled with the use of high-NO_3 groundwater for irrigation has led to increased availability of NO_3 for leaching in the soils (Spalding and Exner, 1993). In view of the above observations, we believe that accounting for the mass of NO_3 in irrigation water in the seasonal crop nutrient requirements can be a potential remedial measure that will also reduce resource requirements for agricultural production in the TRP region.

In addition to agricultural practices, geologic and soil hydrological properties that influence overall groundwater movement and vadose zone processes have major implications on the fate and transport of NO_3 (Nolan et al., 2002; Masetti et al., 2008). The unconsolidated quaternary sand–gravel formations

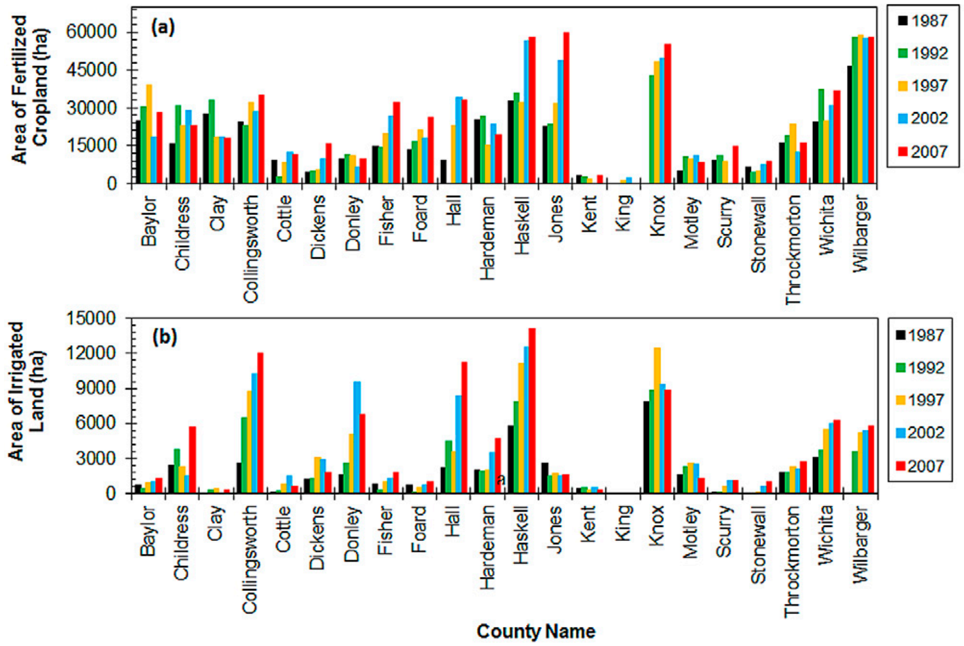

Fig. 8. Area estimates of (a) fertilized crop lands and (b) irrigated lands in counties of the Texas Rolling Plains. (Data source: National Agricultural Statistics Service.)

typically found in the TRP counties are highly transmissive and favor groundwater movement and solute transport through the geologic media. Some of the highest transmissibility and permeability coefficients in this region have been reported from Haskell and Knox Counties (Duffin and Beynon, 1992), indicating better groundwater circulation and thus greater potential for NO_3 movement in these counties. This is supported by the observation that >90% of the total observations in the TRP region that exceeded the NO_3 MCL in the 1990s and the 2000s were from the unconsolidated sandy formations. In contrast, the majority of the observations from the mudstone, shale, or clay/mud formations had low NO_3 concentrations (<20 mg L^{-1} NO_3). Clays, major constituents of shale and/or mudstones, impede groundwater and solute movement due to

Table 2. Descriptive summary of groundwater nitrate concentration in the Texas Rolling Plains Counties in the 2000s.

County	Sample size	Minimum	25th Percentile	Median	75th Percentile	Maximum	SD
		mg L^{-1} as NO_3					
Baylor	2	26.7	NA†	112.1	NA	197.4	120.7
Knox	12	47.8	58.8	80.8	155.6	177.5	49.4
Haskell	18	50.5	57.9	79.5	121.2	160.7	33.4
Wichita	5	38.5	40.3	66	93.2	112.9	29.9
Wilbarger	17	8.3	19.6	50.5	83.7	105.8	33.2
Fisher	6	29.4	30.3	48.7	69.3	85.9	21.4
Collingsworth	39	0	15.8	36.5	60.7	156.3	36.5
Childress	9	3	5.1	31.1	63.8	79.7	30.2
Jones	7	2.3	9.3	22.1	51.4	70.4	24.4
Cottle	14	2.4	7.2	21.7	43.5	88.1	27
Hardeman	12	0.1	12	20.4	44.9	113.3	31.6
Kent	4	2.6	3	20	41.9	43.9	21.4
Hall	5	4	4.7	17.9	46.2	55.3	22
Donley	21	0.8	5.3	14	28.8	65.7	15.7
Foard	7	0.9	4.3	13.1	23.4	86.8	29.7
Stonewall	2	4.6	NA	12.3	NA	20	10.9
Motley	7	3.2	3.9	10.6	95.2	164.2	63.4
King	4	0.2	1.8	9.1	12.7	13.1	5.8
Dickens	4	2.5	3.4	7	40.2	50.9	22.8
Scurry	17	0.1	0.2	4.5	16.2	53.2	18.9
Clay	1	NA	NA	NA	NA	NA	NA
Throckmorton	0	NA	NA	NA	NA	NA	NA

† Not available.

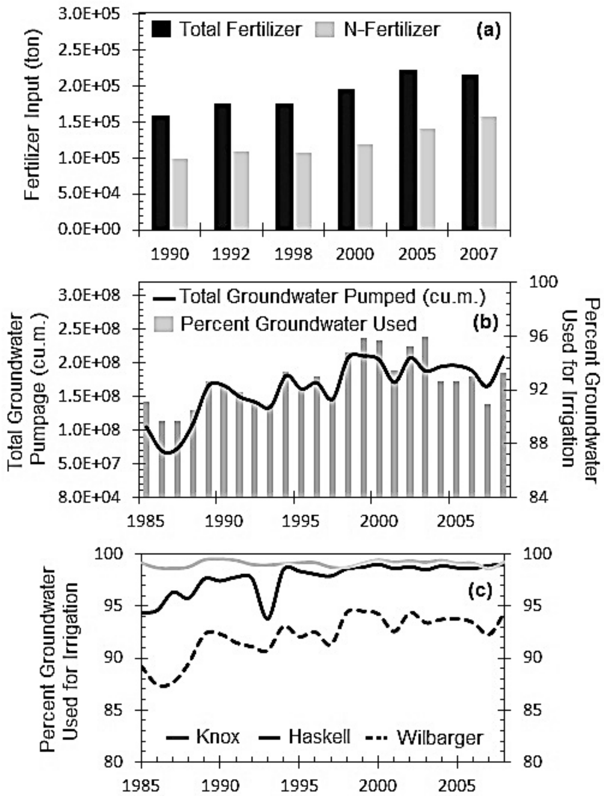

Fig. 9. Temporal pattern of (a) fertilizer input for upland cotton in Texas. Total fertilizer input indicates the sum of nitrogenous, phosphate, and potash fertilizers. (Data source: National Agricultural Statistics Service.) (b) Total pumped groundwater and the percent groundwater used for irrigation in Seymour aquifer and (c) percent groundwater pumped for irrigation in Knox, Haskell, and Wilbarger Counties between 1985 and 2008. (Data source: Texas Water Development Board.)

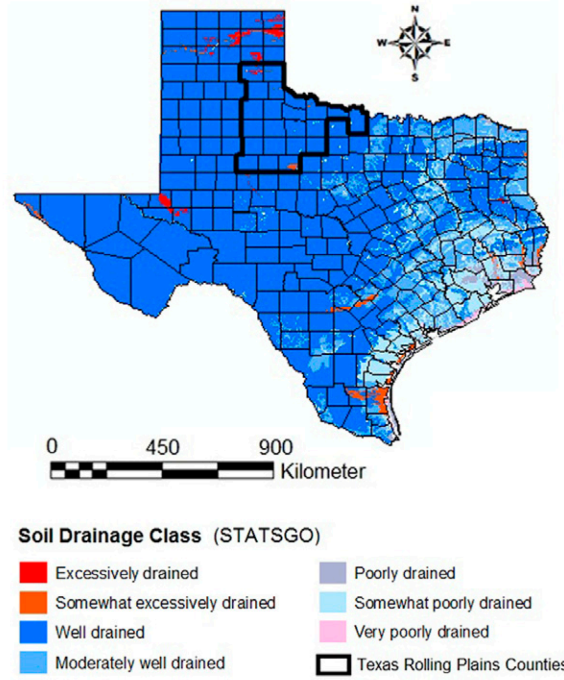

Fig. 10. Statewide distribution of major soil drainage classes. (Data source: State Soil Geographic Database.)

their fine pore size and the lack of interconnected pathways. In addition to geologic characteristics, evaluation of soil hydrologic properties provided important clues to understand the spatial distribution pattern of groundwater NO_3 concentrations across the state. Well drained soils facilitate faster infiltration and thus NO_3 leaching (Costa et al., 2002; Nolan et al., 2002). Soil drainage characteristics revealed that most of west, north, and north-central Texas (including the TRP) is characterized by well drained soils (Fig. 10). These observations indicated that a vast land area of the state has favorable geologic and soil properties for NO_3 leaching. In contrast, eastern Texas counties are characterized by poorly drained soils that restrict water infiltration and NO_3 movement down the soil profile to groundwater systems, which accounted for lower NO_3 concentrations in these counties.

In this study, we used county-based aggregated information on fertilized croplands and irrigated lands due to the lack of availability of adequate spatial information. Soil organic matter, a major driver of nutrient, water, and energy in the ecosystems, was not taken into consideration in the analysis. The soil organic matter is an immense reservoir of organic N and plays a significant role in the N cycling in soil, including N mineralization and nitrification/denitrification. Therefore, in addition to the anthropogenic activities (i.e., agricultural practices) that lead to NO_3 contamination, spatio-temporal analysis of soil N transformations, especially at locations with recognized NO_3 risk, is important to assess groundwater NO_3 contamination patterns. The groundwater quality assessments can further be improved by considering the information on spatial distribution of agricultural activities (fertilizer application rates and timing, irrigation patterns, and tillage operations), climatic variables (precipitation and temperature), and chemical and hydrologic characteristics of the aquifer material (soil and geology). We intend to address these research gaps in our future efforts.

Table 3. Spearman rank correlation between median nitrate concentration and various agricultural parameters in the Texas Rolling Plains.

Time period	Area of fertilized cropland, 2007	Area of irrigated land, 2007	Area of agricultural land, 2006
2000s	0.70***	0.53*	0.74***
	Estimated area of fertilized cropland, 1997	**Estimated area of irrigated land, 1997**	
1990s	0.68***	0.36	
	Estimated area of fertilized cropland, 1987	**Estimated area of irrigated land, 1987**	
1980s	0.53*	0.66**	

* Significant at the 0.05 probability level.

** Significant at the 0.01 probability level.

*** Significant at the 0.001 probability level.

Conclusions

Spatio-temporal assessment of groundwater quality data indicated that the groundwater NO_3 concentrations have increased significantly in several parts of Texas between the 1960s and the 2000s, especially in the TRP region, indicating an immediate need to identify the problem areas and take appropriate preventive and remedial measurements. In the 2000s, there were 25 counties in the state (nine in the TRP) with >30% observations exceeding the NO_3 MCL, as compared with eight counties (five in the TRP) in the 1960s. In the TRP region, about 40% of the observations exceed the NO_3 MCL in the 2000s, compared with about 24% in the 1960s. In Knox and Haskell Counties, both located in the TRP, all observations exceeded the MCL in the 2000s. Groundwater NO_3 concentrations were spatially autocorrelated across the state, with a distinct spatial clustering of high-NO_3 counties observed in the TRP and parts of the SHP in recent times. Spatial clustering pattern of counties appeared to be a temporal phenomenon, with the autocorrelation indices becoming increasingly stronger over time. Our analysis suggested that several factors, such as agricultural activities, geologic characteristics, soil types, and climate, worked in conjunction to influence the spatio-temporal patterns of groundwater NO_3 concentrations in Texas. Although groundwater NO_3 concentrations are increasing in different parts of the state, adequate groundwater quality data to support research and/or decision-making is significantly lacking, as indicated by the lack of NO_3 data from seven counties in the 2000s. As the importance of groundwater as major water source continues to increase in Texas, a more critical review of the groundwater quality and quantity information in different parts of the state will be necessary. Some effort to characterize NO_3 travel pathways through aquifers with different hydrostratigraphic and hydrogeochemical characteristics and land management practices in Texas might be useful to understand the specific interactions between NO_3 and the porous media and the response patterns of different hydrologic systems, which are critical in designing appropriate mitigation and protection strategies.

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