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RESEARCH ARTICLE

Assessing the land use type and environment factors affecting groundwater nitrogen in an arid oasis in northwestern China

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Abstract
Identifying the magnitude and seasonal variability of groundwater nitrogen (N) under various land use types and quantifying the contribution of their environmental factors are of great importance when attempting to implement prioritizing effective strategies for mitigating groundwater N pollution. In this study, hydrochemical investigation was used to assess the magnitude and temporal variability of groundwater N in arid regions. Spatial distributions of N species (total N (TN), nitrate-N (NO_3^-–N), ammonium-N (NH_4^+–N), and nitrous-N (NO_2^-–N)) were mapped using geostatistical techniques. Redundancy analysis (RDA) was conducted to determine environmental factors controlling hydrochemistry. The results showed that residential areas (town and village) and cropland had higher groundwater N concentrations than natural (forest and grassland) and unused land. And the concentrations of N species in rain season (August) were greater than those in the dry season (March) and normal season (November). The N species spatial patterns showed that there is a risk of TN and NO_3^-–N pollution in groundwater of town and surrounding developed cropland, and that NH_4^+–N and NO_2^-–N pollution were negligible. Selected environmental factors explained a total of 77.4% of data variance in N concentrations. These factors indicated that water environmental factors (dissolved oxygen (DO), oxidation–reduction potential (ORP), water temperature (WT), and pH) affect groundwater concentrations and forms of N by influencing the process of nitrification and denitrification, which explained about 60% of the variance of the data. Approximately 10.8 and 8.3% of the variability was explained by shallow groundwater depth and soil texture, indicating that N concentrations in groundwater had heterogeneous influence. The high N excessive pollution ratio was observed in towns and cropland indicating that artificial N input is the main reason for groundwater N pollution in the study area. Hence, ameliorating anthropogenic agricultural practices and reducing N input in urban areas are critical to alleviating groundwater N pollution in the research area.

Keywords Environmental factors · Nitrogen pollution · Redundancy analysis · Temporal–spatial variations · Zhangye Oasis

Introduction

Anthropogenic nitrogen (N) pollution in groundwater is an international issue causing long-term damage on the environment, threatening local ecosystems, economy, and human health; these damages are more pronounced in arid regions (Yang and Liu 2010; Wick et al. 2012; Paredes et al. 2020). In arid regions, surface water resources are almost negligible, and groundwater is the main supplier of water for industrial, agricultural, and domestic uses (Liu et al. 2017; Zhou and Zhi Zhao 2019). Human demand for freshwater resources has increased with a growing population and rising living standards in arid regions, putting enormous pressure on the already scarce water resources (El Alfy et al. 2017). Furthermore, the coexistence of underground aquifer with leaching of fertilizers, industrial or domestic wastewaters, and animal farming wastes has led to irreversible degradation of groundwater quality by N in arid regions, and this problem has raised universal concerns around the world (Chen et al. 2018; Jia et al. 2020).

Groundwater N concentrations are mainly caused by excessive N leaching from the surface; however, groundwater N

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concentrations vary with land use type because of the different N sources (Gu et al. 2013; El Alfy et al. 2017). Previous research has shown that excess N-fertilizers applied to increased agricultural yields is the main source of N pollution in groundwater, and nitrate concentrations of groundwater in agricultural land may be a magnitude higher than those in natural and semi-natural vegetation (Zhang et al. 2013; Yang et al. 2017; El Alfy et al. 2017). N in groundwater may originate from other diffused or point sources, such as industrial sewage, landfill leachate, and sewer leakage, which are of growing importance alongside industrialization and urbanization; therefore, many groundwater N pollution cases were reported beneath urban lands, especially in the emerging developing cities with high-density population and inadequate leak-proof sanitation facilities (Böhlke et al. 2006; Wick et al. 2012; Gu et al. 2013). Different land use types may have different sensitivities to groundwater N magnitude and dynamics owing to the various pollution sources involved. Understanding and quantifying the magnitude and dynamics of N in groundwater under different land use types are critical for implementing source control and alleviating groundwater N pollution.

Moreover, spatiotemporal variation of groundwater N concentrations is usually “specific in situ” even in the same land use because of the various environmental factors. In general, aquifer properties are the primary factor for shallow groundwater N pollution from nitrification and denitrification rate of N species. And, for example, the nitrification rate increases with the increase of dissolved oxygen (DO) content in water, and by contrast, the denitrification rate, however, shows an opposite tendency (Zhang et al. 2016; Chai et al. 2019). Nitrification and denitrification rate were appropriate in a neutral environment (pH value ranges from 6.5 to 8), inhibiting in environment with strong acid (pH < 5.5) and strong base (pH > 9.5) (Zhang et al. 2015a, 2015b); Temperature will affect the growth rate and generation cycle of nitrification and denitrification microorganisms (Hammer and Knight 1994), and the optimum temperature of nitrification rate and denitrification rate ranges from 30 to 35 and 15 to 30 °C, respectively (Andrade and Stigter 2009; Schot and Pieber 2012). Soil physicochemical properties also play an important role in N pollution of groundwater; soil texture controls groundwater N concentrations by regulating N leaching coefficient; groundwater N concentrations in sandy soils are significantly higher than those in loam; and in the same soil texture, that with high organic matter content has a higher N leakage coefficient (Su et al. 2014; Li et al. 2018). Shallow groundwater depth determines the time and distance of soil nitrate leaching; generally, the deeper the groundwater depth, the longer the distance and time of N leaching to groundwater and the lower the probability of groundwater contamination by nitrate (Chen et al. 2018; Li et al. 2018). Understanding the environmental factors of groundwater N concentrations is essential for prioritizing effective strategies and management practices to reduce N pollution.

Zhangye Oasis is an artificial oasis with large population density, and relatively developed industry and agriculture. Zhangye Oasis produces among the highest crop yields with irrigated agriculture. In recent decades, large areas of the surrounding desert have been converted to a variety of productive land use types to meet the growing social and economic needs. However, the sandy desert soil has rough texture, loose structure, and extremely low organic matter content; crop production in such soil must rely on large amounts of fertilizer and irrigation, resulting in high rates of N leaching into the shallow aquifer (Yang and Liu 2010; Su et al. 2014). In addition, industrial sewage, landfill leachate, sewer leakage in urban areas alongside the growing developing industrialization and urbanization, and random discharge of untreated domestic sewage in rural environments increase N pollution of groundwater (Yang and Liu 2010; Gu et al. 2013). Although some researchers have reported groundwater N pollution in the Zhangye Oasis, these studies are limited to agricultural land and lack of comprehensive quantification of N dynamic under different land use types (Yang and Liu 2010; Fang and Ding 2010; Su et al. 2014; Sheng et al. 2019). More importantly, researches involving temporal and spatial variations and influencing factors on groundwater N pollution have seldom been reported in this area.

The main purpose of this paper was to investigate the changes of N species (TN, NO_3^-–N, NH_4^+–N, and NO_2^-–N) under different land use types on the spatiotemporal scale, and the environmental factors influence groundwater N concentrations in Zhangye Oasis. To achieve these goals, we first analyzed N species concentrations in 84 groundwater wells to determine groundwater N variability on temporal scale. Second, we illustrated N species concentrations through inverse distance weighting interpolation method on spatial scale. Third, we evaluated the N species pollution level in groundwater under various land use types. Finally, we determined the relationships between groundwater N concentrations and environmental factors and quantified the interpretation rate of single environmental factor for N variation. The results will guide management strategies which are intended to prevent and control groundwater N pollution.

Materials and methods

Study area

Zhangye Oasis, a national commercial grain demonstration base in Gansu province, is located in the middle reaches of the Heihe River basin. This region has a typical continental climate with an annual precipitation of 117 mm and annual potential pan evaporation of 2390 mm, and 70% of the annual precipitation occurs in summer season (Zhao et al. 2010; Zhang et al. 2017). The main soil types are fluvo-aquic and irrigation-sediment soils distributed in oasis area along the Heihe River and irrigation-farmed sandy and irrigation-farmed gray-brown desert soils on the edge of the oasis (Su et al. 2017). There was sandy land irrigation cropland with different degrees of maturity due to the different cultivation years and characterized by high sand content, low organic matter content, and poor soil water storage capacity (Zhang et al. 2017). Oasis agriculture in this area has a thousand-year history, giving oasis the name of "Gold Zhangye" because of its superior natural environment and highly developed irrigation agriculture. In fact, this area was first in Gansu Province to produce seed corn, sugar beet, and Chinese herbal medicines. Intensive agricultural activity in this region requires large amounts of irrigation and groundwater supplies more than 70% of the local irrigation demand (Mi et al. 2016).

Meanwhile, Zhangye Oasis is a bridge connecting Europe and Asia due to its critical geographic location, has been an important post town on the "Silk Road" since ancient times, and has become an important business circulation, energy base, and transportation hub city in Gansu Province. The urbanization process of Zhangye Oasis is characterized by increasing urban population, growing industrial outputs, and expanding urban built-up area. Accompanied with the implementation of the western development strategy, the administrative level of Zhangye has risen from the county to the city; the population of Zhangye Oasis has expanded significantly, reaching 0.803 million by 2018, and the urbanization level has increased from 21.2% in 2000 to 49.9% in 2018. The total industrial output in Zhangye Oasis reached 10.7 billion Chinese Yuan in 2018 and increased by 8.6 times compared with 2000 (Zhangye Statistical Yearbook 2018). At the end of the twentieth century, the urban built-up area has entered a stage of rapid expansion, increasing from 12.54 km^2 in 2000 to 31.9 km^2 in 2018, with an expansion rate of 1.07 $km^2\ a^{-1}$ (Zhangye Statistical Yearbook 2018). The peak of population occurs in July to August and February to March, which is mainly caused by the influx of tourists in summer and the return of migrant workers for the Chinese New Year (Zhang et al. 2015a, 2015bFeng et al. 2017). There are three industrial parks in Zhangye Oasis, mainly including mining, cement, metallurgy, power plants, and other polluting enterprises, which caused relatively serious industrial point source pollution, such as the contamination of groundwater N (Wen et al. 2009; Fang and Ding 2010; Feng et al. 2017). In order to achieve the goal of environmental protection in the 13th 5-year plan (2016–2020), Zhangye's government carried out a series of rectification on traditional pollution enterprises and successively closed some enterprises that failed to meet the emission standards, and as a consequence, the environmental remediation has achieved initial results.

Land use types

The DEM and land use map of Zhangye Oasis for 2015 were provided by the Heihe Plan Data Management Center (http://westdc.westgis.ac.cn) in the form of WGS-1984-Alber projection with a spatial resolution of 30 m (Fig. 1b). Cropland is the dominant type of land use, covering 68.3% of the total oasis area. Until 2010, the cultivated area was 2896 km^2, with the exception of the old oasis cropland (cultivation period > 80 years) along the Heihe River, and nearly half of the cultivated land was the result of continuous expansion into the desert over the past 50 years (Su et al. 2014; Yang et al. 2017). Approximately 75% of agricultural land was densely cultivated with seed and field maize using traditional methods of plastic film mulching and flood irrigation to maintain yields; further, approximately 200–400 kg N hm^{-2}, 90–150 kg P_2O hm^{-2}, and 60–90 kg K_2O hm^{-2} were applied to fields of maize during cultivation every year (Su et al. 2017). Some cash crops such as greenhouse vegetables, wine grapes, stevia, and Chinese herbal medicines are cultivated by individual families in dispersed plots. The forest is mainly composed of *Elaeagnus angustifolia* and *Populus alba* and xerophytic and super-xerophytic shrubs, such as *Tamarix chinensis*, *Nitraria sphaerocarpa*, and *Reaumuria soongorica*. The grassland contains mainly arid and semi-arid desert types, such as *Agropyron cristatum*, *Kalidium*, and *Phragmites australis*. Unused land refers to land with a vegetation cover below 5%, including sand, saline–alkali land, and bare land. The study area consists of three county-level administrative areas, namely, Gaotai, Linze, and Ganzhou (Fig. 1b). Residents are distributed inside the oasis, with the town in a concentrated distribution pattern, and the rural residents scattered in the oasis; residential areas are connected by intercity highways and country roads.

Sample collection and chemical analysis

We sampled groundwater during 3 distinct seasons: wet in August 2018, normal in November 2018, and dry in March 2019. We collected a total of 252 groundwater samples from 84 wells that included national monitoring, irrigation, and hand-pumping wells in Zhangye Oasis. Collected groundwater samples were placed in polythene bottles and transported to the laboratory for storage at − 20 °C. The location and shallow groundwater depth and land use type were noted. Environmental factors such as DO, ORP, WT, and pH were measured on-site using YSI EXO multi-parameter water quality analyzer (USA) during samples collection. TN, NO_3^-–N, NH_4^+–N, and NO_2^-–N were analyzed using SEAL flow injection AutoAnalyzer 3 (BRAN + LUEBBE, AA3, Germany). Concentrations of NO_3^-–N, NH_4^+–N, and NO_2^-–N were determined after filtration through a 0.45-μm membrane; TN concentrations were determined using the alkaline

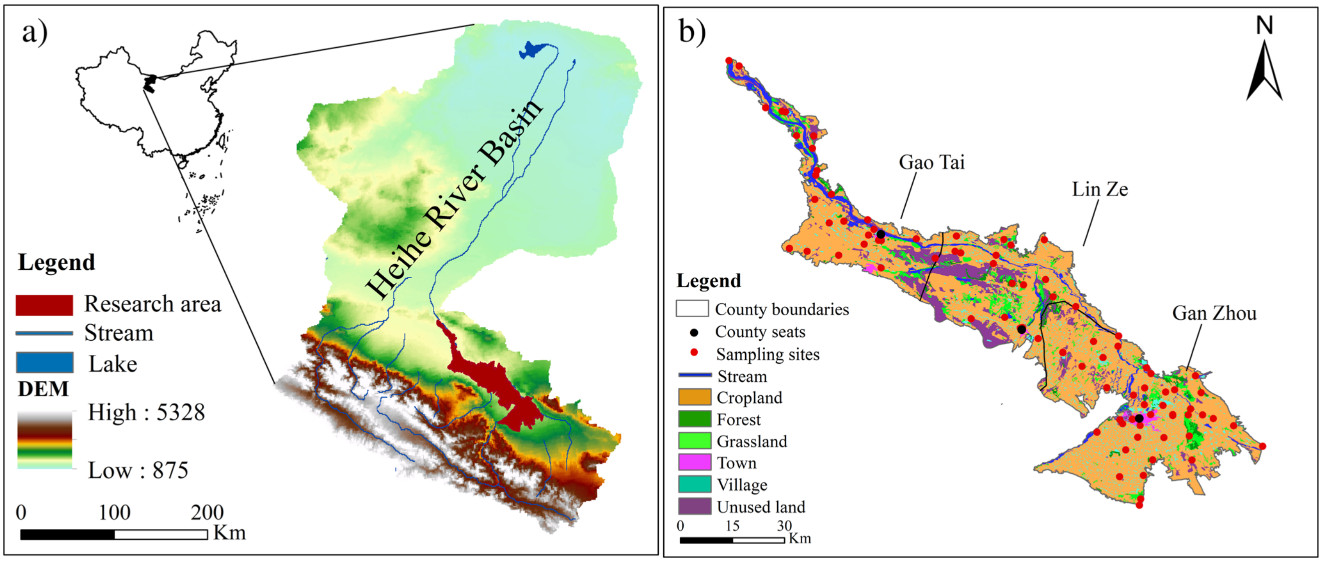

Fig. 1 Location of the study area (**a**) and land use and location of groundwater sampling sites (**b**) in Zhangye Oasis

potassium persulfate ($K_2S_2O_8$) digestion and UV spectrophotometry (Wang et al. 2015).

Data analysis

Statistical and redundancy analyses

Temporal variability in groundwater N concentrations were evaluated with ANOVA (Duncan, $p < 0.05$) of SPSS 21.0 (Chicago, IL, USA). Kolmogorov–Smirnov's test ($p > 0.05$) was used to determine temporal distribution of N-species concentrations in all land use types (Chen et al. 2018). Redundancy analysis (RDA) was used to analyze the ordination relationship between N-species concentrations and their environmental factors, such as DO, ORP, WT, pH, SGL, and ST. The Monte Carlo permutation test (T = 999) was used to detect the significance of effects of environmental factors on the distribution of N concentration. The interpretation ratio of individual environmental factor to total variation in N concentrations was analyzed using the rdacca.hp package in R software. The advantage of the package is that it can partition the interpretation ratio of individual factor interpretation variable of RDA, and a permutation test was used to obtain P value for the ratio with a theory of hierarchical partitioning. RDA ordination, environmental factors interpretation ratio, and the Monte Carlo permutation test were done with the rdacca.hp, vegan, and permu.hp package, respectively, in RStudio 3.5.2 software (http://cran.r-project.org/).

Spatial variability and interpolation methods

Geostatistical methods were used to analyze spatial variability in groundwater N concentrations and assess N concentration level through interpolation method. The semivariogram analysis allows calculation of the experimental semivariance scatter plot first, and then fitting a function to the data points to build a theoretical semivariogram model (Schaefer et al. 2010). Semivariogram analysis of TN, NO_3^-–N, NH_4^+–N, and NO_2^-–N concentrations in groundwater was carried out using GS+ version 9. Spatial variability includes both random and structural variation, where nugget (C_0) represents internal randomness of the data caused by sampling error, short-range variability, and intrinsic variability; partial sill (C) indicates structural variability of the data caused by non-human factors such as topography, climate, soil, and parent material; sill ($C_0 + C$) represents total variance of the data; range represents the scope of spatial autocorrelation; nugget coefficient $C_0/(C_0 + C)$ indicates the strength of spatial heterogeneity, where, generally, nugget coefficient at < 25%, between 25 and 75%, and > 75% indicates strong, moderate, and weak spatial autocorrelation of the data, respectively. The time factor in groundwater TN, NO_3^-–N, NH_4^+–N, and NO_2^-–N concentrations was spatially determined by inverse distance weighting interpolation method (Serio et al. 2018), and spatial representations of N concentrations were done with ArcGIS 10.3 software (ESRI, Redlands, CA, USA).

Results

N concentrations in shallow groundwater

The descriptive statistic results of N concentrations under different land use types are presented in Table 1. The mean and the median concentrations of N species were similar across the land types. Concentration ranges of TN, NO_3^-–N, NO_2^-–N, and NH_4^+–N were 0.5~43.32, 0.02~36.34, 0.01~1.1, and 0.01~0.91 mg L^{-1}, respectively. The mean values of TN are

Table 1 Concentrations of groundwater N species across different land use types

N species	Statistical values	Forest (n = 33)	Grassland (n = 33)	Cropland (n = 90)	Village (n = 33)	Town (n = 42)	Unused land (n = 21)
TN	Range (min–max)	0.7–6.04	0.5–7.03	0.52–32.5	0.95–13.89	1.2–43.32	0.97–10.85
	Mean ± SD	3.3 ± 1.45c	3.78 ± 1.94c	12.64 ± 9.14a	7.58 ± 3.84b	14.11 ± 10.35a	5.81 ± 3.56bc
	Media	3.45	4.36	9.41	7.39	12.75	4.94
	CV	0.44	0.51	0.72	0.51	0.73	0.61
NO_3^-–N	Range (min–max)	0.03–4.18	0.02–4.49	0.06–29.72	0.46–11.95	0.46–36.34	0.31–8.98
	Mean ± SD	2.14 ± 1.05c	2.3 ± 1.35c	10.35 ± 8.25 a	5.6 ± 3.18b	11.41 ± 9.04a	4.11 ± 2.72bc
	Media	2.42	2.62	7.52	5.47	9.57	3.43
	CV	0.49	0.59	0.80	0.57	0.79	0.66
NH_4^+–N	Range (min–max)	0.01–0.56	0.02–0.64	0.01–0.91	0.03–0.36	0.11–0.84	0.01–0.91
	Mean ± SD	0.2 ± 0.15b	0.24 ± 0.16ab	0.3 ± 0.23a	0.19 ± 0.08b	0.21 ± 0.03a	0.12 ± 0.03b
	Media	0.15	0.21	0.22	0.18	0.25	0.15
	CV	0.75	0.67	0.77	0.42	0.14	0.25
NO_2^-–N	Range (min–max)	0.01–0.24	0.02–0.55	0.02–1.1	0.01–1.01	0.1–1.04	0.01–0.44
	Mean ± SD	0.08 ± 0.06d	0.2 ± 0.14 c	0.33 ± 0.24ab	0.36 ± 0.29a	0.37 ± 0.25a	0.23 ± 0.11bc
	Media	0.06	0.16	0.29	0.32	0.30	0.23
	CV	0.75	0.70	0.73	0.81	0.68	0.48

the largest, and those of NO_3^-–N are following, while those of NO_2^-–N and NH_4^+–N are relatively small and have no significant difference among various land use types in Zhangye Oasis. The highest mean concentrations of groundwater TN, NO_3^-–N, and NO_2^-–N were observed in town, at 14.11, 11.41, and 0.37 mg L^{-1}, respectively; maximum groundwater NH_4^+–N concentration was found in cropland at 0.3 mg L^{-1}. The range of CV is 0.14 to 0.8 for N species concentration which indicates an intermediate variability intensity (the CV value between 0.1 and 1.0 represents an intermediate variability intensity) (Sun et al. 2010).

In terms of the significance of various of N concentrations in shallow groundwater wells, TN and NO_3–N concentrations in groundwater wells in cropland and town were significantly different (p < 0.05) with those in all other land use types; however, those in forest, grassland, and unused land did not differ significantly from each other (p > 0.05). There were significant differences (p < 0.05) among NO_2^-–N concentrations in village, town, unused land, forest, and grassland. NH_4^+–N concentrations did not exhibit significant differences among forest, grassland, village, and unused land.

Values in the same row followed by the same letter are not significantly different at P< 0.05

N chemical composition in groundwater (Table 2) showed that NO_3^-–N was the main N species, accounting for 60.9~81.9% of TN concentrations with mean of 78%. Proportions of NH_4^+–N and NO_2^-–N to TN were 2.4~6.5 and 2.6~5.3%, respectively. However, N chemical composition was different in various land use types. Cropland and town were characterized by a higher proportion of NO_3^-–N, accounting for 81.9 and 76.2% of TN, respectively. The highest proportion of NH_4^+–N to TN was found in grassland at 6.5 and 5.3% of TN, respectively

Temporal variability in N concentrations

TN, NO_3^-–N, NH_4^+–N, and NO_2^-–N concentrations in shallow groundwater in all land use types revealed temporal variability between August 2018, November 2018, and March 2018 (Fig. 2). Both the concentrations of NO_3^-–N and NH_4^+–N show an increase tendency from March 2019 to November 2018 and further to August 2018. The range of concentrations of NO_3^-–N is 1.19~9.63, 1.88~11.47, and 2.38~12.57 mg L^{-1} in the three periods, respectively, while those of NH_4^+–N are 0.1~0.23, 0.13~0.28, and 0.2~0.37 mg L^{-1}, respectively. Concentrations of NO_2^-–N showed the trend of the highest in August 2018, while the difference between March 2019 and November 2018 was not significant. TN and NO_3^-–N concentrations differed significantly (p < 0.05) within a single hydrological year (August 2018 to March 2019) between cropland, village, and town and forest and grassland; however, NH_4^+–N concentrations did not differ (p > 0.05) across various land use types. NO_2^-–N concentrations were not significantly different (p > 0.05) in cropland, village, and town but were in the forest (p < 0.05). Generally, groundwater N concentrations were relatively stable over time in forests, grassland, and in unused land; in contrast, N concentration varied greatly over time in cropland, village, and town.

Table 2 Proportions of NO_3^-–N, NH_4^+–N, and NO_2^-–N to TN in various land use types

N species	Forest	Grassland	Cropland	Village	Town	Unused land	Mean
NO_3^-–N	65.1	60.9	81.9	73.0	76.2	71.8	78.0
NO_2^-–N	3.1	5.3	2.6	4.7	2.6	3.9	3.1
NH_4^+–N	6.2	6.5	2.4	2.5	6.3	3.3	2.8

Spatial variability in N concentrations

Spatial structure in groundwater N concentrations

The results of the semivariogram model (Table 3) showed that the nugget coefficients for groundwater TN, NO_3^-–N, NH_4^+–N, and NO_2^-–N concentrations were 2.9, 16.67, 5.42, and 4.89%, respectively, suggesting that N-species concentrations in shallow groundwater had strong spatial autocorrelation, and that structural factors affected groundwater N spatial variability, in that, spatial variability of NO_2^-–N was more random, and that of NO_3^-–N was least random among N-species. The spatial autocorrelation ranges of TN, NO_3^-–N, NO_2^-–N, and NH_4^+–N concentrations in monitoring wells were 420, 400, 660, and 760 m, respectively, indicating that groundwater N was distributed continuously in this spatial range.

Spatial distribution of groundwater N concentrations

Groundwater in the study area was classified into I~V water quality categories according to guidelines for drinking water quality, 4th edition, by the World Health Organization (WHO) (Fig. 3; Table 4). The threshold values for pollution from NO_3^-–N, NO_2^-–N, and NH_4^+–N concentrations were 10, 1, and 0.5 mg L^{-1}, respectively. TN standard was not set by WHO, and NO_3^-–N concentrations accounted for 76% of TN concentrations in shallow groundwater in the study area;

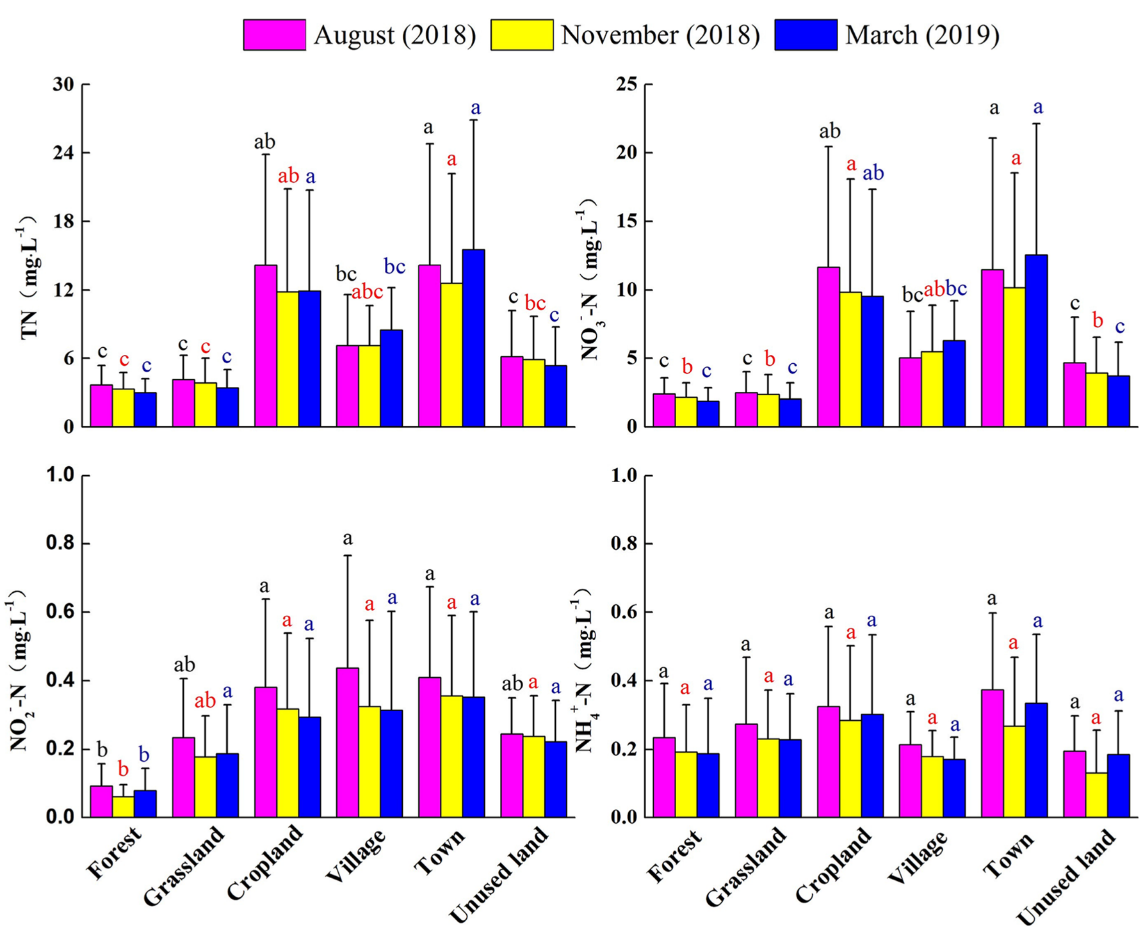

Fig. 2 Temporal variability in N concentrations in shallow groundwater in Zhangye Oasis

Table 3 Semivariance model and its parameters for N species concentrations

N species	Model type	Nugget (C_0)	Sill ($C_0 + C$)	Range/m (A)	$C_0/(C_0 + C)$/ %	R^2	Residual
TN	Spherica	0.03	1.01	420	2.9	0.22	0.167
NO_3^-–N	Gaussian	0.011	0.066	400	16.67	0.044	0.089
NO_2^-–N	Spherica	0.011	0.225	660	4.89	0.236	0.127
NH_4^+–N	Spherica	0.0098	0.164	760	5.42	0.133	0.022

therefore, we classified TN concentrations according to the standards for NO_3^-–N concentrations.

Median values of TN and NO_3^-–N concentrations in shallow groundwater ranged from 5.0 to 20.0 mg L^{-1} and were found in 84 and 68.3% of the area of Zhangye Oasis, respectively (Table 4). Using the WHO threshold values for NO_3^-–N pollution (10 mg L^{-1}), TN and NO_3^-–N pollution in groundwater was present in 34.0 and 20.4% of the study area, respectively, indicating the occurrence of TN and NO_3^-–N pollution. Most of the NH_4^+–N concentrations were in the III level water quality (0.1–0.5 mg L^{-1}), and NH_4^+–N pollution existed in about 3.2% of Zhangye Oasis. A large portion of Zhangye Oasis exhibited NO_2^-–N concentrations between 0.1 and 0.5 mgL^{-1}, indicating low risk of NO_2^-–N pollution.

The N concentrations in shallow groundwater displayed different variations under the various land use types (Fig. 4).

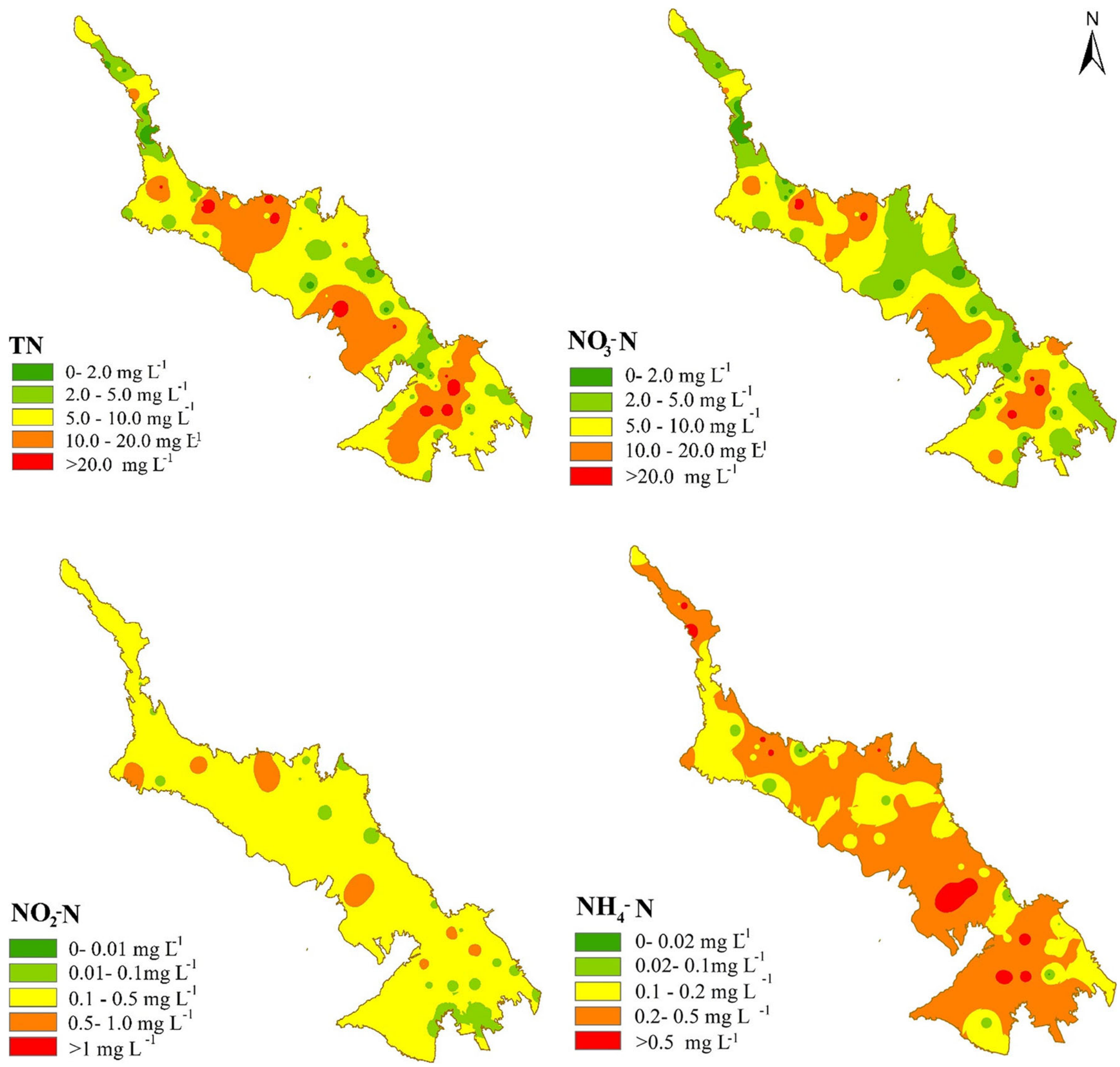

Fig. 3 Spatial distribution of median concentrations of groundwater N species in Zhangye Oasis using inverse distance weighting interpolation method ($n = 84$)

Table 4 The area ratio for different levels of N species concentrations in shallow groundwater (%)

N species	I	II	III	IV	V
Range	0–2 mg L^{-1}	2–5 mg L^{-1}	5–10 mg L^{-1}	10–20 mg L^{-1}	> 20 mg L^{-1}
TN	1.3	12.6	52.2	31.8	2.2
NO_3^-–N	2.4	28.5	48.6	19.7	0.7
Range	0–0.02 mg L^{-1}	0.02–0.1 mg L^{-1}	0.1–0.2 mg L^{-1}	0.2–0.5 mg L^{-1}	> 0.5 mg L^{-1}
NH_4^+–N	0	1.6	29.2	66	3.2
Range	0–0.01 mg L^{-1}	0.01–0.1 mg L^{-1}	0.1–0.5 mg L^{-1}	0.5–1.0 mg L^{-1}	> 1 mg L^{-1}
NO_2^-–N	0	5.4	89.3	5.3	0

Groundwater sample pollution frequency (percentage of total samples over 10 mg L^{-1}) for TN and NO_3^-–N was 57.1 and 43.3% in town and 50 and 40% in cropland. And there was no groundwater sample that exceeded pollution threshold of TN and NO_3^-–N (10 mg L^{-1}) in forest and grassland. Except for 7.1% of groundwater samples from town indicated NO_2^-–N pollution, and there was no pollution risk in our research area. The vast majority of NO_2^-–N concentrations in most land use types were categorized at level III, and 88% of groundwater samples in forest were at level II. There was a low-level NH_4^+–N pollution in the study area, and approximately 20, 13, 10, and 10% of the NH_4^+–N samples exceeded the threshold of 0.5 mg L^{-1} in cropland, town, forest, and grassland, respectively.

Relationship between groundwater N concentrations and environmental factors

The RDA results indicated that both the model and the ordination axis reached a significant level ($p < 0.05$) with the Monte Carlo permutation test (T = 999) (Table 5). The sum of the eigenvalues of the first two axes was 0.816 and cumulatively explained 99.22% of the spatial distribution of groundwater N concentrations. The correlation coefficient between groundwater N concentrations and environmental factors for axis 1 and axis 2 were 0.936 and 0.342, respectively. Those results suggested that the ordination results can better explain the relationship between environmental factors and N concentration distribution. We selected the first two ordination

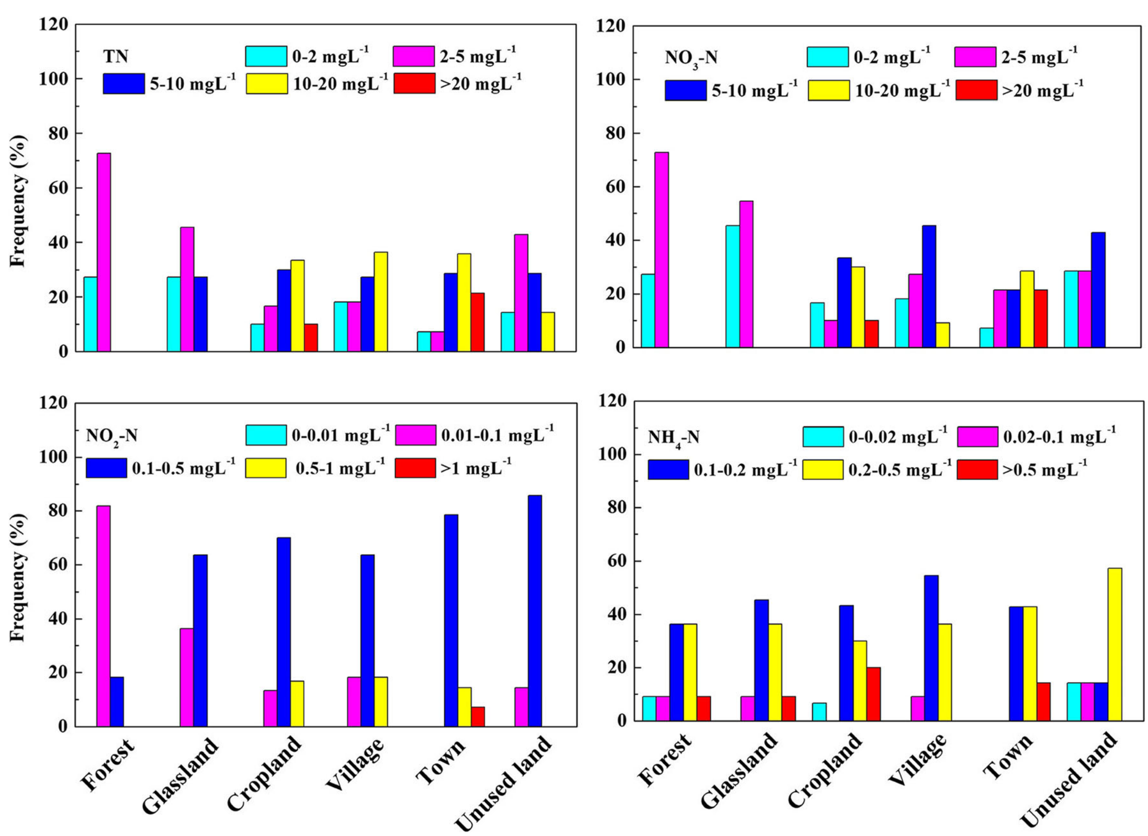

Fig. 4 Frequency distribution of N species concentrations in shallow groundwater samples in Zhangye Oasis ($n = 252$)

Table 5 RDA ordination summary for groundwater N concentrations in Zhangye Oasis and influencing factors

Axes	1	2	3	4
Eigenvalue	0.7968	0.0191	0.0064	0.000
Correlation of N concentration factors	0.936	0.342	0.233	0.201
Cumulative proportion of N concentrations (%)	96.90	99.22	99.99	100
Monte Carlo permutation test of model	$R^2 = 0.822$	$P = 0.001$***	F = 31.28	

axes to draw a two-dimensional RDA ordination diagram (Fig. 5).

The ordination diagram (Fig. 5) between groundwater N concentrations and environmental factors showed that groundwater concentrations of TN and NO_3^-–N were negatively correlated with DO and pH and significantly positively correlated with ORP and ST. In addition, TN concentrations were significantly positively correlated with NO_3^-–N concentrations in groundwater. NH_4^+–N concentrations were significantly positively correlated with WT, and NO_2^-–N concentrations were significantly positively correlated with SGL and ORP. Moreover, there was a significant correlation between environmental factors; ORP was positively correlated with SGL, and DO was significantly negatively correlated with ORP. There were significant differences in groundwater N concentrations among different land use types. Groundwater N concentrations in forest and grassland were mainly distributed in the second and third quadrants, in village and town—in the first and fourth quadrants—and in cropland and unused land—in all four quadrants (Fig. 5). This indicated that N concentrations were highly variable.

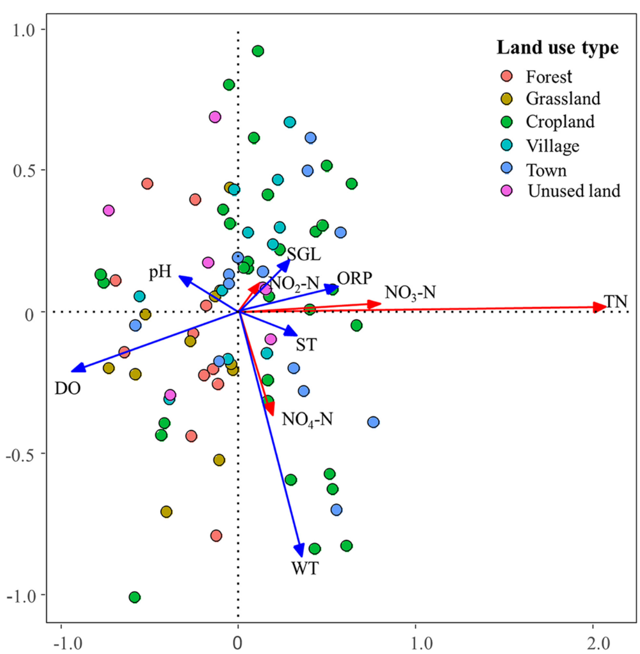

Fig. 5 RDA ordination diagram of shallow groundwater N concentrations and environmental factors

The interpretation ratio of the selected environmental factors for the total variation in N concentrations was 77.4%, and 22.6% of the total variation was not explained (Fig. 6). DO exhibited the maximum interpretation ratio of 38.5% for the distribution of groundwater N concentrations, followed by ORP (13.9%), SGL (10.8%), ST (8.3%), WT (3.9%), and pH (2%) (Fig. 6a). The proportion of explained variation by an individual influence factor to total was consistent with the proportion of total variation with a maximum interpretation ratio by DO and a minimum interpretation ratio by pH. Influencing factors, such as DO, ORP, SGL, and ST, significantly affected groundwater N concentrations ($p < 0.05$), while WT and pH did not ($p > 0.05$). In fact, water-related environment factors (e.g., DO, ORP, WT, and pH) were the main influencing factors of groundwater N concentrations in Zhangye Oasis, and they explained nearly 60% of the variability in N concentrations. Approximately 10.8 and 8.3%of the variability in N concentrations was explained by SGL and ST, respectively.

DO, ORP, WT, and pH are dissolved oxygen, oxidation–reduction potential, water temperature, and pH of shallow groundwater, respectively. SGL is shallow groundwater depth, and ST is soil texture.

Discussion

N species and groundwater quality

In this study, NO_3^-–N was the main N species in groundwater in Zhangye Oasis. The highest ratio of NO_3^-–N to TN was observed in cropland, followed by residential area (town and village), unused and natural land (grassland and forest) (Table 2). Possible reasons to a high proportion of NO_3^-–N concentrations in agro-ecosystem may be agricultural practices, increasing application of fertilizers to improve crop yields, mineralization of urea, decomposition of ammonium nitrate, and organic fertilizers, which are stored in the soil in the form of NO_3^-–N, and leak into groundwater with irrigation and rainfall (Kaushal et al. 2011; Ma et al. 2019). Other anthropogenic N sources, such as sewage effluent, landfill leachate, and leaking manure, can be dissociated by anaerobic denitrifying bacteria into NO_3^-–N stored in groundwater; this may account for high NO_3^-–N ratio in residential areas (Gu

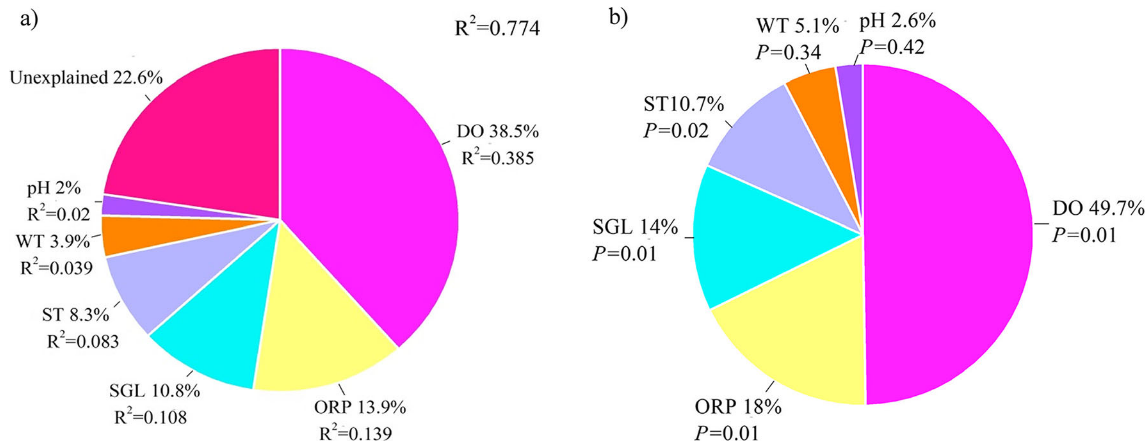

Fig. 6 Proportions of total variation (**a**) and total explained variation (**b**) explained by individual environmental factors.

et al. 2013; Wu and Sun 2016). Moreover, forms of N in groundwater are also affected by land use. Jiao et al. (2017) pointed out that the percentage of NO_3^-–N in TN was 46.7 and 72.8% in forest and tea plantation, respectively. Chen et al. (2018) found the ratio of NO_3^-–N to TN to be as high as 84.6% beneath vegetable fields. These results are consistent with our finding that the ratio of NO_3^-–N to TN in an agro-ecosystem was significantly higher than that in a natural ecosystem.

The soil has an open and stable framework with network pores and high specific surface area, and these structures make the soil have a negative charge and a high cation exchange capacity (Zhang et al. 2016). The negative charge within the soil pores can be physically adsorbed by some cations such as Na^+, Ca^{2+}, and Cu^{2+}, and the high cation exchange capacity allows these cations to be considerably exchanged with some cations in soil, such as ammonium ions (Komarowski and Yu 1997; Rozic and Cerjan-stefanovic 2000). Therefore, NH_4^+–N is often adsorbed on the soil surface, making it hard to leak into the aquifer. Our research found that NH_4^+–N contamination was detected in approximately 3.2% of the area in Zhangye Oasis (Fig. 3); these results are consistent with the findings of 13 regions in Saudi Arabia (Alabdulaaly et al. 2010), the Ningxia irrigation area in northwest China (Chen et al. 2018), and Fasarud Plain in southern Iran (Bahrami et al. 2020). However, Wang et al. (2015) noted that NH_4^+–N accounted for 45–50% of TN concentrations in paddy fields and double-rice fields, showing a moderate NH_4^+–N pollution level in subtropical central China. Zhao et al. (2016) reported that > 70% of groundwater sampling sites exceeded the threshold of 0.5 mg L^{-1}, and NH_4^+–N was the dominant N pollutant in Dongting Lake, China. Two explanations may account for the differences in these research results: (1) the latter two studies were located in a subtropical region with abundant precipitation, where urea can be rapidly hydrolyzed into NH_4^+–N during downward leakage with water flow, and (2) the aerated zone of soil turns into a reducing environment in flooded conditions, which inhibits growth of aerobic nitrifying bacteria, limits nitrification, and promotes storage of NH_4^+–N in groundwater (Krupa et al. 2011; Wang et al. 2015).

Influences of land use on groundwater N pollution

Land use type has a direct impact on groundwater N concentrations; changes in land use affect surface cover and interfere with soil physical and chemical properties and microbial activity resulting in N migrating from the soil surface to the groundwater hydrological circulation system, aggravating groundwater N pollution (Liu et al. 2014; Wang et al. 2015; Zhao et al., 2016; Lasagna et al. 2016). In our study, the rank of NO_3^-–N concentration in different land uses is town > cropland > village > unused land > grassland > forest, and the majority of samples with groundwater NO_3^-–N concentrations exceed 10 mg L^{-1}, from residential areas (town and village) and cropland (Fig. 4), indicating human activities had a considerable effect on groundwater N concentrations.

Population growth, urban expansion, and socioeconomic development all intensively affect the magnitude and dynamics of regional N in groundwater (Grimm et al. 2008). Gu et al. (2013) reported NO_3^-–N in groundwater of China from1980 to 2010 finding that although cropland still is the main source of N leaching to groundwater, industrial waste, sewer leakage, and landfill leachate are the fastest-growing factors contributing to groundwater N concentrations alongside the rapid urbanization and industrialization. Accompanied with the implementation of the development of the western region in China, Zhangye's government had introduced a large number of enterprises to develop the local economy, among which are some polluting enterprises such as metallurgy, coal, and power station etc. (Fang and Ding 2010; Feng et al. 2017). There was more than 1000 tons of industrial N that are discharged into the Heihe River every year, and as consequence, plenty of N enters groundwater under the effect of water cycle; this maybe the most important reason for the highest NO_3^-–N concentrations that occurred in town (Zhangye City Ecological Environment Bureau 2018). Meanwhile, the lack of urban sewage treatment facilities may be another reason for the significant N pollution of groundwater in residential areas. In 2018, there was a total of 34.44 million tons of domestic sewage produced by town in Zhangye Oasis, of which approximately 8.34 million tons are directly discharged into the environment; moreover, only 34% of the villages are equipped with sewage network pipes to centrally treat domestic sewage, and the majority of domestic waste is directly buried in the soil by individual families (Zhangye City Ecological Environment Bureau 2018).

Several studies reported that excessive use of agricultural N fertilizer was the main source of N in agricultural ecosystems (Kaushal et al. 2011; Wang et al. 2015). N fertilizer application rate in Zhangye Oasis area is generally as high as 200–400 kg N hm^{-2} $year^{-1}$; only 35–45% is absorbed by plants; and 10–15% would enter into groundwater, contributing a high NO_3^-–N concentration in cropland (Su et al. 2014). Irrigation may be another factor in higher NO_3^-–N concentrations in cropland in arid regions. Assouline et al. (2015) reported that irrigation was a dominant driving force for leachate of N species from soil to underground system in arid regions, and multiple re-circulation of irrigation groundwater would further aggravate NO_3^-–N pollution. In our research area, more than 70% of irrigation water comes from groundwater; corn is irrigated about 8~10 times; and vegetables approximately 8~14 times during the growing season, the annual irrigation amount averages 1200 mm in the cropland, of which about 39% will leak into the 200-mm soil layer forming deep drainage, and ineffective irrigation scheduling is one of the key factors of groundwater pollution in oasis cropland (Zhang et al. 2018; Zhang et al. 2019). Irrigation methods can also intensively affect the groundwater N concentrations. In our study, more than 80% of the cropland use traditional methods of flood irrigation, and the mean of NO_3^-–N in cropland was 10.35 mg L^{-1}, significantly higher than the cropland with advanced irrigation method of dropper in arid region, such as Yanqi Basin, Xinjiang (3.32 mg L^{-1}) (Wang et al. 2016), Saudi Arabia (4.39 mg L^{-1}) (Abdel-Satar et al. 2017), and Kerman Province in Iran (4.63 mg L^{-1}) (Moosavirad et al. 2013). These practices resulted in a significant groundwater NO_3^-–N pollution in agricultural lands in Zhangye Oasis.

In contrast, groundwater in natural land (forest and grassland) had the lowest N concentrations. Zhang et al. (2013) found that groundwater NO_3^-–N in agro-ecosystems may be one magnitude higher than that in natural land in China. Human activities can affect groundwater N concentrations in natural land via elevated N deposition (Liu et al. 2013). Using the lowest threshold value of 3 mg L^{-1} NO_3^-–N in N deposition (Andrade and Stigter 2009) shows little incidence of anthropogenic groundwater NO_3^-–N pollution in natural areas. However, due to dry climate, rare precipitation events, and underdeveloped industry and agriculture in arid regions of northwestern China, N deposition rate is 0.02–0.73 g N m^{-2} a^{-1}, or only 22% of that in the North Plain, and 34% of that in southern China (Gu et al. 2016). Because groundwater N was low in natural lands in our study area, we conclude that the effect of atmospheric N deposition on groundwater N in natural land in Zhangye Oasis is negligible.

We detected a significant seasonal pattern in groundwater N concentrations in different land use types. N concentrations were almost the same in normal (November) and in dry season (March), and significantly lower in wet season (August). That was mainly due to precipitation and irrigation, with > 70% of annual rainfall occurring in summer; moreover, crop growth requires fertilization and irrigation in August (growth period), resulting in N leaching to the groundwater system in summer, especially in agro-ecosystems (Su et al. 2014; Mi et al. 2016). However, groundwater NO_3^-–N concentrations in residential areas (town and village) showed an opposite trend, with NO_3^-–N concentrations higher in dry season than in wet season. That was due to high population density in the dry season in Zhangye Oasis. Namely, migrant work is popular in Zhangye Oasis; workers return to towns and villages for the New Year, leading to increased sewer generation and leakage into groundwater system (Zhang et al. 2015a, 2015b Feng et al. 2017).

Factors affecting N concentrations in shallow groundwater

In our study, the selected environmental factors explained 77.4% of the variability, and dissolved oxygen (DO) (P = 0.01), oxidation–reduction potential (ORP) (P = 0.01), shallow groundwater depth (SGL) (P = 0.01), and soil texture (ST) (P = 0.02) were the most significant factors contributing to the variability in groundwater N concentrations (Fig. 6). DO and ORP are critical parameters of oxidation–reduction reactions in shallow groundwater. DO content controls the rate of nitrification and denitrification, and ORP determines oxidation–reduction conditions (Chai et al. 2019). In general, organic N in wastewater and soil, including protein, amino acid, amide, and urea, is mineralized to ammonia. Ammonia is soluble in water and can be ionized into ammonium ions in aerobic conditions. Aerobic microorganisms convert ammonium ions to nitrate via nitrification, and nitrate can persist in an anaerobic environment (Yen et al. 1996; Böhlke et al. 2006). However, nitrification is inhibited when DO in the groundwater system is < 2 mg L^{-1} (Maltais-Landry et al. 2009). In our study, concentrations of DO in shallow groundwater ranged from 1.84 to 7.18 mg L^{-1}, indicating that DO can provide satisfactory levels of oxygen for nitrification, continuously oxidizing NH_4^+–N into NO_3^-–N in the shallow groundwater system. The ORP is below 0 mV, considering a restoration environment, where NO_3^-–N has high stability (Maltais-Landry et al. 2009). The ORP was between − 447 and − 304 mV in this study, indicating that NO_3^-–N was more stable, and remaining as the main form of TN in groundwater (Chen et al. 2018). Some studies also reported that DO and ORP had notable effects on groundwater N species concentrations (Yen et al. 1996; Zhao et al., 2016). However, Almasri (Almasri and Kaluarachchi 2004) found that there was no active decrease of NO_3^-–N concentrations when DO was sufficient in the shallow groundwater system. This shows that the effect of DO on the form of groundwater N is also controlled by other factors, which still need to be examined.

The depth of groundwater determines the vertical migration distance and time for N leaching, thus regulating N leaching rate. In general, the deeper the shallow groundwater, the lower the risk of groundwater N pollution (Yang et al. 2017). This phenomenon can be explained by nitrification–denitrification and groundwater vertical movement (Almasri and Kaluarachchi 2004). NO_3^-–N is stable in water and can be easily transported into deeper groundwater systems with precipitation or irrigation. In soils with large soil particles and good air permeability, oxygen is sufficient, beneficial to nitrification; however, once the soil aeration zone is saturated with moisture, the environment becomes anaerobic and reducing, favoring denitrification (Lasagna et al. 2016; Zhao et al., 2016; Chen et al. 2018). The rate of nitrification and denitrification may change at different depths, in 0–4 m soil layer; nitrification dominates under sufficient oxygen conditions, with more NH_4^+–N oxidizing to NO_3^-–N; NO_3^-–N concentration increases with shallow groundwater depth. As groundwater depth increases, oxygen needed by nitrifying bacteria decreases, denitrifying bacteria become more active and dominant, and NO_3^-–N is further oxidized to N_2, and returned to the atmosphere (Liu et al. 2005; Lasagna et al. 2016).

Soil texture controls groundwater N concentrations by regulating the N-leaching coefficient, and soil particle size determines soil water holding capacity and nutrient transport, which in turn affects N leaching. In general, soil with high clay content has relatively high nutrient retention capacity, which can prevent some N from leaching; in contrast, N in sandy soils with unobstructed drainage and weak water holding capacity is more likely to follow water movement to groundwater (Su et al. 2014). Simmelsgaard (1998) reported that when the average N application rate is 168 kg·$(hm^2\ a)^{-1}$, and clay particle content in soil at 0–25 cm depth is 5, 12, and 20%, leaching losses of N are 68, 44, and 26 kg·$(hm^2\ a)^{-1}$. Su et al. (2014) and Li et al. (2018) also reported that groundwater N concentrations in sandy soils are significantly higher than those in loam. The above results are consistent with the results of our study; with an increase in soil clay particle content, the leaching losses of N decreased. Water temperature and pH affected N the concentrations in shallow groundwater by influencing nitrification and denitrification process (Zhao et al., 2016); these were weakly correlated with concentrations of N of various N forms in our study.

Conclusions

This paper uses an integrated approach involving hydrochemical investigations, geostatistical techniques, and redundant analysis to analyze the groundwater N dynamics and its relationship with environmental factors in Zhangye Oasis. Hydrochemical analysis shows that groundwater N concentrations vary with seasons; TN and NO_3^-–N concentrations in wet season were higher than in dry season, and this is mainly related to N leaching caused by summer rainfall and cropland irrigation. In contrast, NH_4^+–N and NO_2^-–N fluctuated slightly in various land use types. NO_3^-–N was the main N form in groundwater, and NO_3^-–N > 10 mg L^{-1} was present in 20.4% of the total area of Zhangye Oasis, and mainly concentrated in residential areas and surrounding developed cropland, so strengthening the construction of residential sewage treatment facilities and improving cropland management measures are the main ways to improve groundwater N pollution in the research area. The results of the semivariance analysis show that the shallow groundwater TN, NO_3^-–N, NH_4^+–N, and NO_2^-–N in the study area all have strong spatial autocorrelation; the partial sill of N species was less than 25%, indicating that the spatial heterogeneity of N contents in the study area was dominated by systemic variation. Redundant analysis reveals that the selected environmental factors explain 77.4% of the total data various. The findings of this study will guide management strategies intended to assess complex aquifer contamination processes and prevent and control groundwater pollution in arid areas.

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