Archives of Environmental Contamination and Toxicology (2019) 77:79–87
https://doi.org/10.1007/s00244-019-00636-7

Severe Nitrate Pollution and Health Risks of Coastal Aquifer Simultaneously Influenced by Saltwater Intrusion and Intensive Anthropogenic Activities

Jun Wu[1] · Jian Lu[2] · Xiaohu Wen[3] · Zhenhua Zhang[1] · Yichen Lin[2]

Received: 12 February 2019 / Accepted: 23 April 2019 / Published online: 4 May 2019

Abstract
Groundwater quality is critical for regional sustainability and human well-beings in coastal regions, because groundwater is an important water resource for these areas facing water scarcity. Anthropogenic activities might induce nitrate pollution, whereas saltwater intrusion could decrease coastal groundwater discharge into sea to subsequently cause the persistent accumulation of pollutants in coastal aquifer. Rare information is available on the nitrate pollution of coastal aquifer under simultaneous influences of saltwater intrusion and intensive anthropogenic activities. This study investigated the distribution, pollution, possible sources, and potential health risks of groundwater nitrate of typical coastal aquifer simultaneously influenced by saltwater intrusion and intensive anthropogenic activities. The average/maximal concentration of groundwater nitrate was 173.70/824.80 mg/L, indicating the severe accumulation of nitrate in the coastal aquifer. Concentrations of nitrate in coastal groundwater were much higher than those in adjacent seawater. Groundwater salinization did not have significant effects on nitrate distribution. Groundwater in 87.6% of sampling sites was not suitable for drinking based on nitrate evaluation criterion. Anthropogenic activities might induce nitrate pollution in approximately 94.7% of sampling sites. Sources, including sewage and manure, soil nitrogen, and ammonium fertilizers, contributed to groundwater nitrate with concentration > 100 mg/L in the study area, whereas sewage and manure were the predominant source affecting groundwater nitrate in 97.5% of sampling sites. Groundwater nitrate exerted unacceptable noncancer health risks for infants, children, teenagers, and adults in more than 87.6% of the study area. Infants and children were the most susceptibly influenced by groundwater nitrate. It is urgent to take effective measures for controlling groundwater nitrate pollution in the study area.

Nitrate, ubiquitous in various aquatic environments, often exists in groundwater with relatively high concentrations to make groundwater unsuitable for drinking (Chica-Olmo et al. 2014; Stuart et al. 2011; Wen et al. 2018). Excessive nitrate in groundwater for drinking can induce various health concerns, including abortions, blue baby syndrome, increased risks of methemoglobinemia and gastric cancer, damage to stomach lining, mouth ulceration, non-Hodgkin's lymphoma, and reproductive toxicity (Paladino et al. 2018; Rivett et al. 2008; Wu et al. 2018). Therefore, groundwater nitrate deserves more attention.

Groundwater is very important for many regions, including coastal zones with problems of water scarcity (Ketabchi et al. 2016; Wen et al. 2019). Groundwater pollution has become a critical obstacle for regional sustainability and ecological stability (Chica-Olmo et al. 2014; Güler et al. 2013; Wen et al. 2019). Nitrate pollution of groundwater has attracted wide attention in recent years due to its frequent occurrence (Chica-Olmo et al. 2014; Serio et al. 2018; Wu et al. 2018). Approximately 40% of people in China live in coastal zones (Lu et al. 2018), implying that unexpected negative consequences might be induced if groundwater nitrate pollution occurs in these areas, especially the groundwater-dependent regions. Source apportionment of groundwater

✉ Jian Lu
jlu@yic.ac.cn

1 School of Resources and Environmental Engineering, Ludong University, Yantai 264025, Shandong, People's Republic of China

2 CAS Key Laboratory of Coastal Environmental Processes and Ecological Remediation, Yantai Institute of Coastal Zone Research (YIC), Chinese Academy of Sciences (CAS), Yantai 264003, Shandong, People's Republic of China

3 Key Laboratory of Ecohydrology of Inland River Basin, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences, Lanzhou 730000, Gansu, People's Republic of China

nitrate is critical for pollution management and control (Wen et al. 2018; Xue et al. 2009). Dual-isotope method has been widely used to identify the possible sources of groundwater nitrate (Yang et al. 2013; Wen et al. 2018; Xue et al. 2009), because this approach can provide more comprehensive information on groundwater nitrate pollution. Therefore, dual-isotope method is promising for exploring possible groundwater nitrate sources.

Salinization is another problem possibly affecting groundwater quality, especially in coastal zones (Wen et al. 2019). Negative influence of salinization on groundwater quality has been reported (Walraevens et al. 2015; Wen et al. 2019). The coastal aquifers often are suffering from saltwater intrusion (SWI) to cause the loss of groundwater resources (Bear et al. 1999). Additionally, seawater intrusion could decrease the groundwater discharge into sea to possibly cause the high retention time and persistent accumulation of pollutants. Therefore, research work needs to be performed to clarify whether simultaneous saltwater intrusion and intensive anthropogenic activities will induce the nitrate pollution of coastal aquifer. This study performed field sampling, laboratory analysis, and risk assessment to investigate the pollution, possible sources, and potential health risks of nitrate in typical coastal groundwater of China. The objectives of this study were to provide the comprehensive information on nitrate in coastal aquifer under simultaneous influences of saltwater intrusion and intensive anthropogenic activities and to put a basis for groundwater nitrate control and management in coastal zone.

Materials and Methods

Study Area and Chemical Analysis

The study area lies in the coastal zone of Laizhou Bay, which is a typical coastal bay in the northern Shandong Peninsula of China. Saltwater intrusion in the coastal aquifer of Laizhou Bay coastal area has become the most severe in China due to the groundwater overextraction caused by the extremely intensive anthropogenic activities (Wen et al. 2019). The study area, sampling period, and sampling strategy were the same as those previously reported (Wen et al. 2019). Based on Shandong Statistical Yearbook, the population of the study area reached approximately 5 million in 2017 and the nonagricultural population accounted for less than 15%. The quality-centralized water is not supplied for 60% of residents, because most villages are randomly distributed in the study area. Agriculture is the major anthropogenic activity in the study area so that fertilizers and manure also are widely used in this area. Recently, industry and wastewater reuse have been in rapid development in the study area due to the rapid urbanization. Groundwater samples were collected from 113 shallow wells at the yards of the residents, filtered by membrane filters (pore size of 0.45 µm, Pall Life Sciences, MI), and stored at 4 °C until chemical analysis.

An Ultrameter II™ 6P (Myron L Company, USA) was used to measure pH, total dissolved solids (TDS), and electrical conductivity (EC) of groundwater in situ. Concentrations of these parameters were shown by the previous report (Wen et al. 2019). Dionex ion chromatograph (ICS3000, USA) was used to determine the concentrations of major ions, including Mg^{2+}, Ca^{2+}, Na^+, K^+, Cl^-, SO_4^{2-}, and NO_3^-. Titration method was employed to measure the concentrations of HCO_3^-. The compositions of stable isotopes $\delta^{15}N–NO_3^-$ and $\delta^{18}O–NO_3^-$ were determined at the University of California Facility for Isotope Ratio Mass Spectrometry. In total 40 of 113 groundwater samples with nitrate concentration > 100 mg/L were further selected for analysis of dual isotopes.

Nitrate Pollution Evaluation and Source Apportionment of Coastal Groundwater

Groundwater nitrate pollution was evaluated according to the standard for groundwater quality (GB/T 14848-2017) as well as criteria proposed by other scientists (Chica-Olmo et al. 2014; Anayah and Almasri 2009). Evaluation criterion presented by Anayah and Almasri (2009) focuses on anthropogenic influences. Ranking criteria for groundwater quality based on nitrate are shown in Table 1.

Principal component analysis (PCA) also was used to determine preliminarily the possible sources of major ions in groundwater. Possible nitrate sources were determined by dual-isotope method using $\delta^{15}N–NO_3^-$ and $\delta^{18}O–NO_3^-$ values of different nitrate sources. The detailed information on isotopic values of various sources referred to the published articles (Wen et al. 2018; Xue et al. 2009).

Health Risks Assessment

Nitrate in groundwater exerts potential health risks through dermal contact and oral intake. Nitrate is generally regarded to pose noncancer risks (Zhai et al. 2017). Therefore, hazard quotient (HQ) was used to estimate the potential noncancer risks of groundwater nitrate. HQ through oral intake (HQ_{oral}) and dermal contact (HQ_{dermal}) were calculated by the following equations (Wen et al. 2019; Zhai et al. 2017).

$$HQ_{oral} = \frac{C \times OI \times EF_o \times ED_o}{BW \times AT \times RfD}$$

$$HQ_{dermal} = K_P \times C \times CF \times t_{event} \times \frac{EV \times ED_d \times EF_d \times SA}{BW \times AT} \times \frac{1}{RfD \times GIABS}$$

Table 1 Ranking criteria of pollution and intensity of anthropogenic disturbance

Objective	Level	Classification	Value	References
Groundwater quality evaluation	Good for all uses	I	CN ≤ 2.0 mg/L	Standard for groundwater quality (GB/T 14848-2017)
	Good for all uses	II	2.0 mg/L < CN ≤ 5.0 mg/L	
	Suitable for drinking	III	5.0 mg/L < CN ≤ 20.0 mg/L	
	Suitable for agriculture; treatment needed before drinking	IV	20.0 mg/L < CN ≤ 30.0 mg/L	
	Not suitable for drinking	V	CN > 30.0 mg/L	
Groundwater quality evaluation	Good quality	I	C < 37.5 mg/L	Chica-Olmo et al. 2014
	Intermediate quality	II	37.5 mg/L ≤ C < 50 mg/L	
	Poor quality	III	C ≥ 50 mg/L	
Anthropogenic disturbance evaluation	Most likely background concentration	I	0 < C < 5 mg/L	Anayah and Almasri 2009
	Possible human influence	II	5 ≤ C < 15 mg/L	
	Pollution due to human influence	III	15 ≤ C < 50 mg/L	
	Pollution due to excessive human influence	IV	C ≥ 50 mg/L	

CN means concentration of NO_3^-–N; C refers to nitrate concentration

$$HQ = HQ_{oral} + HQ_{dermal}$$

where C refers to nitrate concentration of coastal groundwater; OI means water consumption rate; ED_o represents ingestion exposure duration; EF_o stands for ingestion exposure frequency; AT refers to average lifespan; BW means body weight; RfD represents the reference nitrate dose through oral exposure route; GIABS stands for the fraction of nitrate absorbed in gastrointestinal tract; K_P means dermal permeability coefficient of nitrate; CF means unit conversion factor; t_{event} refers to event duration; EV means the event frequency; ED_d represents dermal contact exposure duration; EF_d refers to dermal contact exposure frequency; SA means human skin surface area. The values of these parameters are listed in Table 2.

Data Processing

All data regarding concentrations, groundwater pollution levels, and health risks posed by nitrate were processed by Surfer 11 (Golden Software LLC, Colorado, USA). PCA was performed by SPSS 19.0 (IBM, New York, USA). The relationship among major ions, pH, TDS, and EC, in groundwater was evaluated by Pearson matrix with Pearson correlation coefficients obtained by SPSS 19.0.

Results and Discussion

Distribution of Major Ions in Coastal Groundwater

The groundwater in the study area was alkaline except the pH of groundwater collected from 6 sites less than 7.0, whereas concentrations of TDS and EC were in the range of 246.79–3858.03 mg/L and 490.00–6332.00 µS/cm, respectively (Wen et al. 2019).

The distribution of groundwater major ions in the study area exhibited significant spatial variations (Fig. 1). Concentrations of Ca^{2+} ranging from 36.94 to 617.94 mg/L with the mean value of 197.99 mg/L were higher than those of the other major cations, whereas concentrations of K^+ varying from BDL (below the detection) to 66.43 mg/L with the average value of 3.86 mg/L were significantly lower than those of the other major cations in groundwater (Fig. 1). Na^+ with high concentrations mainly existed in southwestern and northeastern parts of the study area, whereas Ca^{2+} and Mg^{2+} with high concentrations mainly existed in the southwestern regions. Different from the other major cations, K^+ with high concentrations occurred in the central part of the study area. Concentrations of Cl^- ranged from BDL to 1507.61 mg/L, whereas those of SO_4^{2-} were in the range of BDL-1310.15 mg/L (Fig. 1). Concentrations of HCO_3^- ranged from 59.71 to 541.88 mg/L with the mean value of 256.91 mg/L (Fig. 1). Sites with high concentrations of SO_4^{2-}, Cl^-, and HCO_3^- were mainly distributed in the southwestern regions of the study area, similar to those with high concentrations of Na^+, Mg^{2+}, and Ca^{2+}. Nitrate concentrations varied from 0.59 to 824.80 mg/L with the mean value of 173.70 mg/L (Fig. 1). Average nitrate concentration of coastal groundwater in the study was almost 150–270 times higher than nitrate concentration of adjacent seawater. The highest nitrate concentration occurred near the central region along sea-land interface, which was different from the distribution patterns of the other anions.

A previous study showed that groundwater salinization occurred in the southwestern plains of the study area, and heavy metal pollution was affected by salinization (Wen et al.

Table 2 Parameters used for human health risk assessment

Parameter	Unit	Infants	Children	TF	TM	AF	AM	References
DR (water ingestion rate)	L/d	0.65	1.5	2	2	2	2	Wu et al. 2016; Zhai et al. 2017
EF*i* (ingestion exposure frequency)	days/year	365	365	365	365	365	365	Wu et al. 2016
EF_d (dermal exposure frequency)	days/year	150	150	150	150	150	150	Akhbarizadeh et al. 2016
ED_i (ingestion exposure duration)	year	1	6	18	18	70	70	USEPA 2011; Yang et al. 2018; Zhai et al. 2017
ED_d (dermal exposure duration)	year	0.5	6	8	8	30	30	USEPA 2011; Yang et al. 2018; Zhai et al. 2017
BW (body weight)	kg	7.79	19.73	47.64	51.24	55.18	63.29	Zhai et al. 2017
AT (average lifespan)	days	182.5	2190	6570	6570	25,550	25,550	USEPA 2011
RfD_O (oral reference dose)	mg/(kg·day)	1.6	1.6	1.6	1.6	1.6	1.6	Anornu et al. 2017
EV (event frequency)	event/day	1	1	1	1	1	1	Yang et al. 2018
SA (skin surface area)	cm^2	3416.0	9035.2	14,321	14,321	18,182	18,182	Wu et al. 2016; Zhai et al. 2017
K_P (dermal permeability coefficient)	cm/h	0.001	0.001	0.001	0.001	0.001	0.001	Zhai et al. 2017
t_{event} (event duration)	h/event	1	1	1	1	1	1	USEPA 2004
CF (unit conversion factor)	L/cm^3	0.001	0.001	0.001	0.001	0.001	0.001	USEPA 2004
GIABS (absorption fraction in GIT)	unitless	1	1	1	1	1	1	USEPA 2004
C_W (pollutant concentration)	mg/L	Measured	Measured	Measured	Measured	Measured	Measured	This study

GIT gastrointestinal tract; TF, TM, AF, and AM refer to female teenagers, male teenagers, female adults, and male adults, respectively

Ages of target humans are < 1 (infants), 1–11 (children), 12–19 (teenagers), and 20–70 (adults)

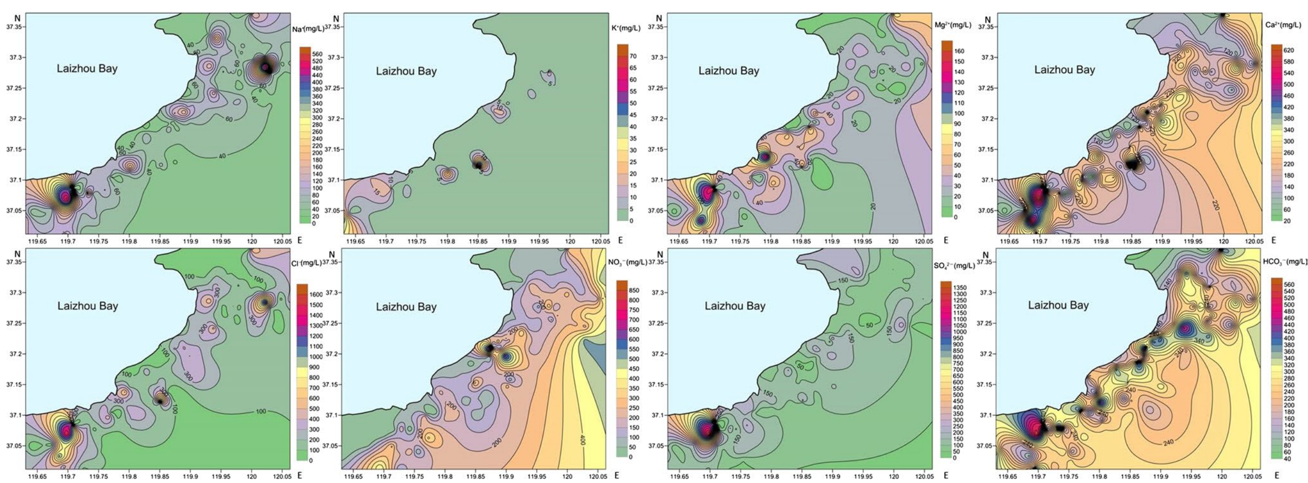

Fig. 1 Distribution of major ions in coastal groundwater

2019). Interestingly, groundwater salinization did not have significant effects on nitrate distribution. A previous study reported that ammonium was easy to accumulate, whereas nitrate showed slight depletion under influences of groundwater salinization (Russak et al. 2015). Therefore, salinization might negatively affect the accumulation of nitrate in groundwater, which explained that high-concentration nitrate did not occur in the salinization regions. Additionally, nitrate concentrations of coastal groundwater in the study area showed different patterns with groundwater salinization, indicating that multiple factors besides groundwater salinization caused by the saltwater intrusion had important impacts on the persistent accumulation of nitrate in the coastal aquifer.

Groundwater Nitrate Pollution in the Study Area

Based on the thresholds of sulfate in groundwater (GB/T 14848-2017), the groundwater quality of 8/75/24 sites was suitable for drinking and categorized into Class I/II/III, whereas that of 4/2 sites was not suitable for drinking and categorized into Class IV/V. Groundwater quality of 21 and 19 sites was categorized into Class IV and V based on criterion of Cl^- standard, whiereas that of the remaining sites was evaluated as Class III or better.

Different criteria were employed to evaluate groundwater nitrate pollution in the study area (Fig. 2). Based on the thresholds of groundwater NO_3^-–N (GB/T 14848-2017), groundwater quality of 3/5/21 sampling sites was suitable for drinking and categorized into Class I/II/III, whereas that of 19.5%/54.9% of sampling sites was Class IV/V (Fig. 2a). Severe nitrate groundwater pollution occurred in the study area, much more serious than that induced by heavy metals (Wen et al. 2019).

Groundwater in the study area was directly used as drinking water. Concentration of 50 mg/L, the recommended maximum allowable value for nitrate in drinking water according to the WHO, was widely accepted threshold for nitrate in drinking water and Chica-Olmo et al. (2014) further refined the evaluation criterion. Based on the criterion proposed by Chica-Olmo et al. (2014), groundwater quality of 12, 2, and 99 sampling sites was determined as good, intermediate, and poor levels, respectively (Fig. 2b). This result showed that groundwater in approximately 87.6% of the study area was not suitable for drinking, more serious than results obtained using Standard for groundwater quality (GB/T 14848-2017). Human activities could have significant impacts on groundwater nitrate (Anayah and Almasri 2009; Wen et al. 2018). The regions with the serious nitrate pollution in the study area were strongly affected by anthropogenic activities (Fig. 2c), showing the great contribution of anthropogenic disturbance to nitrate pollution of coastal groundwater.

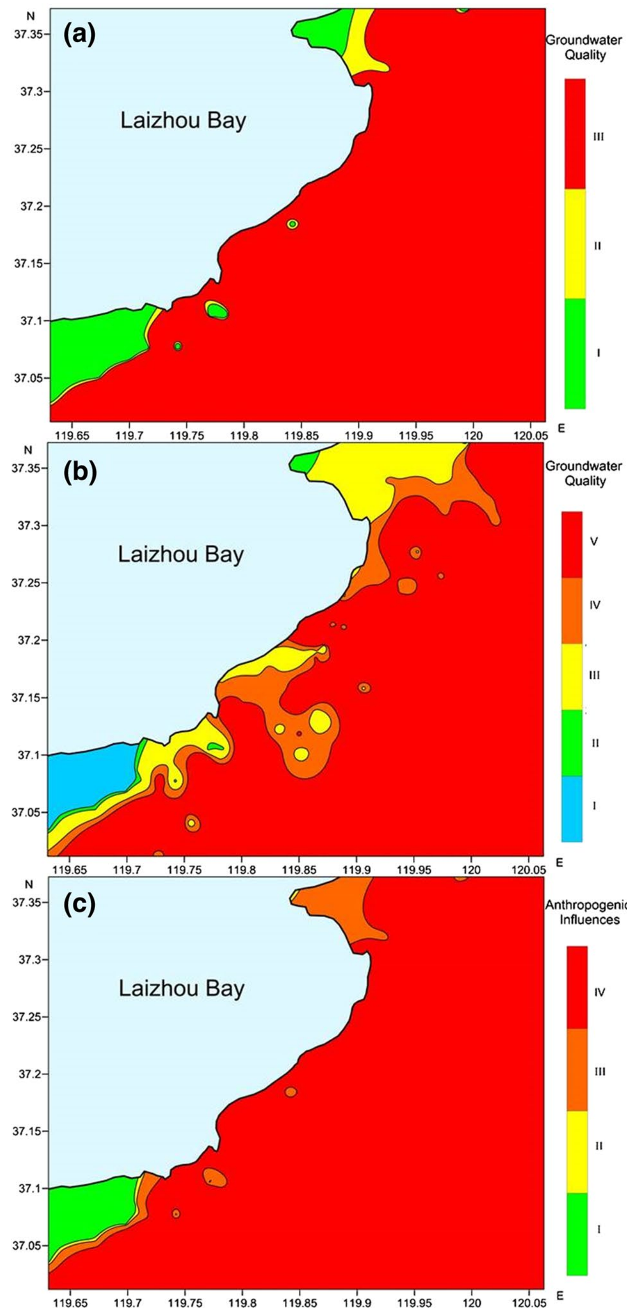

Fig. 2 Nitrate-based groundwater quality of the study area using **a** criterion proposed by Chica-Olmo et al. (2014), **b** standard for groundwater quality (GB/T 14848-2017), and **c** intensity of anthropogenic disturbance

Source Apportionment of Nitrate in Coastal Groundwater

Strong correlation existed among major ions (Table 3). Cations Ca^{2+} and Mg^{2+} were positively correlated with the other major ions at significance level of $p < 0.01$ or $p < 0.05$. Cations (Na^+ and K^+) and anions (SO_4^{2-} and Cl^-) were significantly positively correlated with the other major ions except with NO_3^-. HCO_3^-, TDS, and EC were positively correlated with the other major ions while pH was negatively correlated with the other major ions, TDS, and EC. Interestingly, no significant correlation existed in K^+–NO_3^-, Na^+–NO_3^-, SO_4^{2-}–NO_3^-, and Cl^-–NO_3^-, showing that source of nitrate might be different from that of the other major ions in coastal groundwater.

PCA was used to explore the possible sources of the major ions in coastal groundwater (Table 4). Approximately 69.448% of the total variance was explained by 2 components with eigenvalue > 1.0. The first component consisting of Na^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, and HCO_3^- accounted

Table 3 Pearson correlation of major ions in groundwater

	Na^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	HCO_3^-	TDS	pH	EC
Na^+	1.000										
K^+	0.241*	1.000									
Mg^{2+}	0.538**	0.273**	1.000								
Ca^{2+}	0.567**	0.340**	0.770**	1.000							
Cl^-	0.790**	0.413**	0.749**	0.855**	1.000						
NO_3^-	0.031	−0.130	0.218*	0.443**	0.108	1.000					
SO_4^{2-}	0.722**	0.260**	0.597**	0.569**	0.671**	−0.056	1.000				
HCO_3^-	0.590**	0.240*	0.401**	0.479**	0.396**	0.282**	0.424**	1.000			
TDS	0.801**	0.326**	0.786**	0.922**	0.925**	0.394**	0.726**	0.575**	1.000		
pH	−0.325**	−0.151	−0.313**	−0.553**	−0.413**	−0.323**	−0.316**	−0.405**	−0.497**	1.000	
EC	0.713**	0.346**	0.777**	0.871**	0.886**	0.204*	0.730**	0.389**	0.895**	−0.535**	1.000

*$p < 0.05$; **$p < 0.01$

Table 4 Component matrix and total variance explained for major ions in coastal groundwater

Component	Initial eigenvalues			Elements	Component	
	Total	% of variance	Cumulative %		1	2
1	4.285	53.566	53.566	Na^+	0.832	−0.175
2	1.271	15.882	69.448	K^+	0.437	−0.434
3	0.840	10.502	79.949	Mg^{2+}	0.827	0.090
4	0.748	9.349	89.299	Ca^{2+}	0.884	0.267
5	0.352	4.396	93.695	Cl^-	0.920	−0.111
6	0.306	3.826	97.521	NO_3^-	0.237	0.919
7	0.169	2.109	99.629	SO_4^{2-}	0.788	−0.277
8	0.030	0.371	100.000	HCO_3^-	0.644	0.199

for the largest variance of 53.566%. The second component explaining 15.822% of variance only included NO_3^-. PCA results also proved that source of nitrate in coastal groundwater of the study area was different from that of the other major ions and human activities might be the main nitrate source.

Dual-isotope method was used to identify the possible sources of nitrate in coastal groundwater (Fig. 3). Based on the evaluation criterion (Wen et al. 2018; Xue et al. 2009), groundwater nitrate in approximately 35% of 40 sampling sites with nitrate concentration > 100 mg/L mainly originated from sewage and manure, whereas that in 11 sites were primarily originated from soil nitrogen (N) as well as sewage and manure (Fig. 3). Groundwater nitrate in approximately 20% of target sampling sites mainly came from sewage and manure, soil N, and ammonium fertilizer, whereas that in approximately 15% of sites originated from ammonium fertilizer, soil N, and sewage (Fig. 3). Interestingly, groundwater nitrate of only one target sampling site was mainly influenced by soil N and ammonium fertilizer. Sewage and manure served as the predominant source for nitrate in coastal groundwater of the study area, influencing groundwater nitrate in 97.5% of sampling sites. Therefore, it is critical to control effectively discharge of sewage and manure as well as improve sewage treatment in village areas for reducing groundwater nitrate.

Health Risk Assessment of Nitrate in Coastal Groundwater

Although nitrate was reported to link with cancer (Paladino et al. 2018), cancer slope factor was not proposed and widely accepted for estimating potential cancer risks posed by nitrate. Therefore, this study used hazard quotients to evaluate the potential noncancer risks of groundwater nitrate. Noncancer health risks of groundwater nitrate for four target human groups, including infants (< 1 year old), children (2–11 years old), teenagers (12–19 years old), and adults (20–70 years old), were evaluated (Fig. 4).

HQs of nitrate ranged from 0.012 for male adults at site with nitrate concentration of 0.59 mg/L to 86.12 for infants at site with nitrate concentration of 824.80 mg/L. HQs of nitrate for infants were the highest while those for male adults were the lowest, which was similar to previously report (Zhai et al. 2017). HQs of nitrate for infants were 2.19, 3.97, 4.27, 4.60, and 5.28 times those for

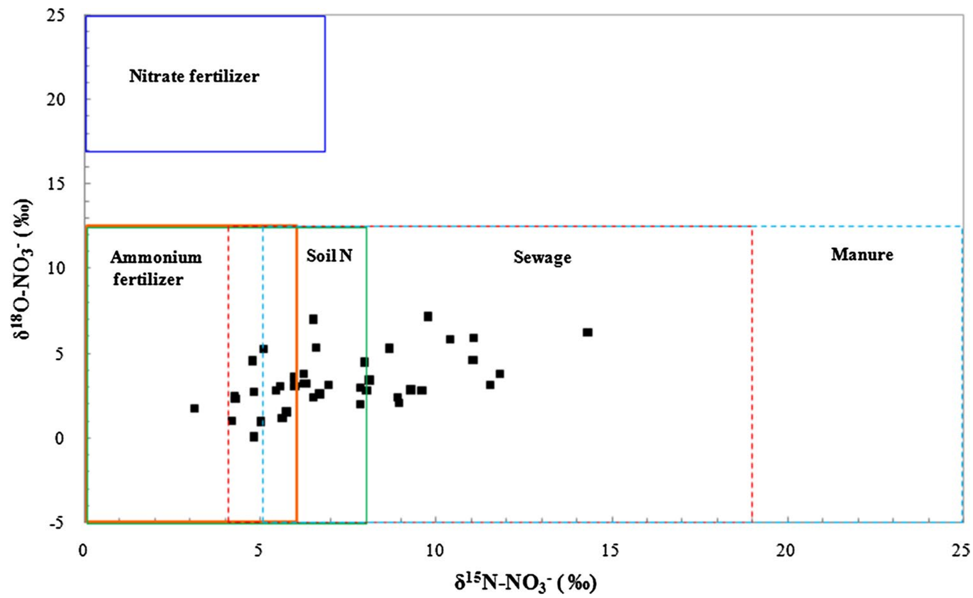

Fig. 3 Source identification of nitrate in coastal groundwater using dual isotopes

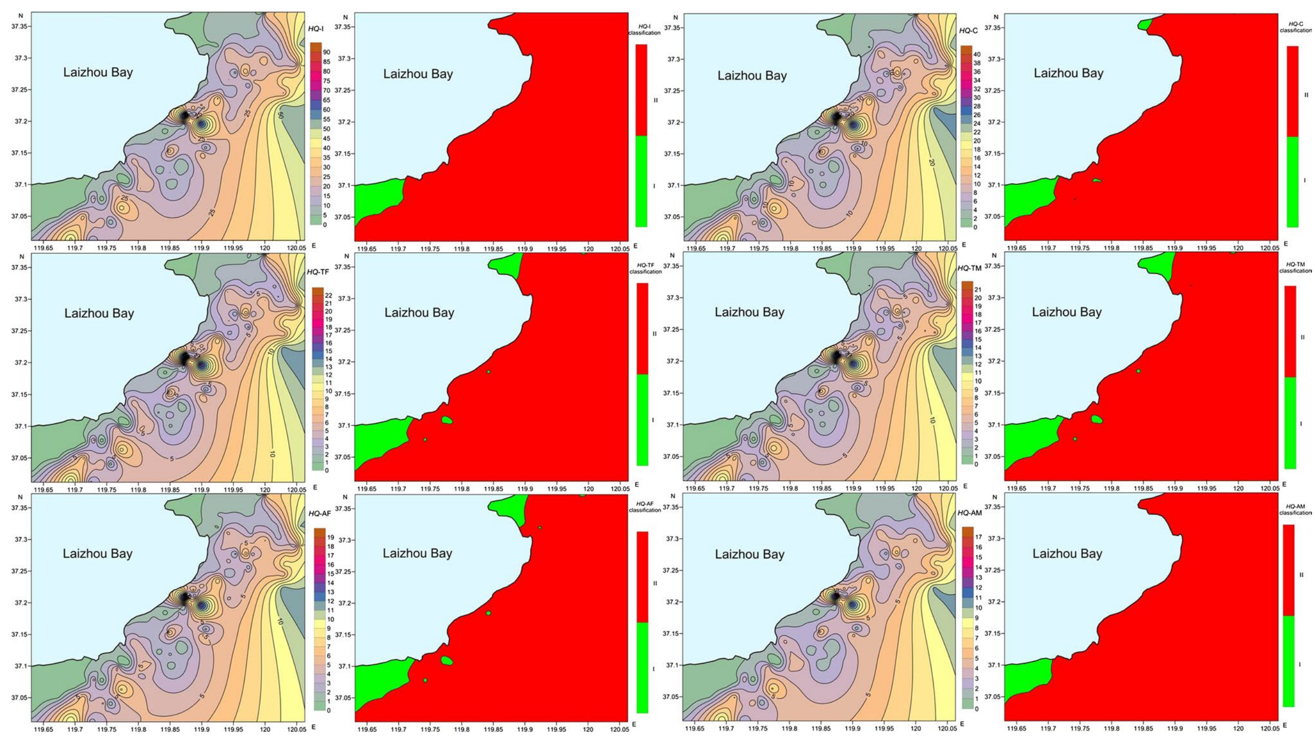

Fig. 4 Noncancer health risks of nitrate in coastal groundwater. HQ means hazard quotient; I, C, TF, TM, AF, and AM refer to infants, children, female teenagers, male teenagers, female adults, and male adults, respectively. HQ levels I and II refer to acceptable and unacceptable health risks, respectively

children, female teenagers, male teenagers, female adults, and male adults, respectively. Oral intake was the main pathway for nitrate exposure to account for approximately 99.75–99.89% of total noncancer risks, similar to previous report on heavy metals (Wen et al. 2019).

Noncancer risks were regarded as unacceptable when HQs > 1 (Anornu et al. 2017; Wen et al. 2019; Zhai et al. 2017). Noncancer risks of nitrate in 96.5%, 92.9%, 89.4%, 87.6%, 87.6%, and 87.6% of the sampling sites were unacceptable for infants, children, female teenagers, male teenagers, female adults, and male adults, respectively. In summary, nitrate with high concentrations in coastal groundwater of the study area would exert serious health risks to the residents, especially infants and children. Therefore, more effective policies should be put forward to control groundwater nitrate pollution of the study area.

Conclusions

The maximal/mean concentration of the nitrate in this area was 824.80/173.70 mg/L, far beyond the WHO drinking water standard. Nitrate concentrations of coastal groundwater in the study area were much higher than those in adjacent seawater, indicating that coastal groundwater might be potential nitrate pollution source for seawater due to possible submarine groundwater discharge. Nitrate concentrations of coastal groundwater in the study area showed different patterns with groundwater salinization, confirming that multiple factors besides the groundwater salinization influenced the persistent accumulation of nitrate in the coastal aquifer. Nitrate pollution in the study area was serious with groundwater in 87.6% of sites not suitable for drinking. Nitrate pollution in approximately 94.7% of sites might be influenced by human activities. Sewage, manure, and fertilizer were the predominant source for high-concentration coastal groundwater nitrate of the study area. Noncancer health risks of groundwater nitrate in 87.6% of the study area were unacceptable for adults, teenagers, children, and infants. Infants were the most influenced group, followed by children. Effective control measures should be proposed to relieve groundwater nitrate pollution and consequent health risks in the study area.

Acknowledgements This work was supported by National Natural Science Foundation of China (41671319), One Hundred Talents Program of Chinese Academy of Sciences (Y629041021), Taishan Scholar Program of Shandong Province (No. tsqn201812116), Two-Hundred Talents Plan of Yantai (Y739011021), and CAS "Light of West China Program." The authors thank the reviewers for their valuable suggestions and comments on the manuscript.

References

Akhbarizadeh R, Moore F, Keshavarzi B, Moeinpour A (2016) Aliphatic and polycyclic aromatic hydrocarbons risk assessment in coastal water and sediments of Khark Island, SW Iran. Mar Pollut Bull 108:33–45

Anayah FM, Almasri MN (2009) Trends and occurrences of nitrate in the groundwater of the West Bank, Palestine. Appl Geogr 29:588–601

Anornu G, Gibrilla A, Adomako D (2017) Tracking nitrate sources in groundwater and associated health risk for rural communities in the White Volta River basin of Ghana using isotopic approach ($\delta^{15}N$, $\delta^{18}O–NO_3$ and ^3H). Sci Total Environ 603–604:687–698

Bear J, Cheng AH, Sorek S, Quazar D, Herrera I (1999) Seawater intrusion in coastal aquifers, concepts, methods and practices. Kluwer Academic Publisher, Dordrecht. ISBN 0-7923-5573-3

Chica-Olmo M, Luque-Espinar JA, Rodriguez-Galiano V, Pardo-Igúzquiza E, Chica-Rivas L (2014) Categorical Indicator Kriging for assessing the risk of groundwater nitrate pollution: the case of Vega de Granada aquifer (SE Spain). Sci Total Environ 470–471:229–239

Güler C, Kurt MA, Korkut RN (2013) Assessment of groundwater vulnerability to nonpoint source pollution in a Mediterranean coastal zone (Mersin, Turkey) under conflicting land use practices. Ocean Coast Manag 71:141–152

Ketabchi H, Mahmoodzadeh D, Ataie-Ashtiani B, Simmons CT (2016) Sea-level rise impacts on seawater intrusion in coastal aquifers: review and integration. J Hydrol 535:235–255

Lu J, Wu J, Zhang C, Zhang Y, Lin Y, Luo Y (2018) Occurrence, distribution, and ecological-health risks of selected antibiotics in coastal waters along the coastline of China. Sci Total Environ 644:1469–1476

Paladino O, Seyedsalehi M, Massabò M (2018) Probabilistic risk assessment of nitrate groundwater contamination from greenhouses in Albenga plain (Liguria, Italy) using lysimeters. Sci Total Environ 634:427–438

Rivett MO, Buss SR, Morgan P, Smith JWN, Bemment CD (2008) Nitrate attenuation in groundwater: a review of biogeochemical controlling processes. Water Res 42:4215–4232

Russak A, Yechieli Y, Herut B, Lazar B, Sivan O (2015) The effect of salinization and freshening events in coastal aquifers on nutrient characteristics as deduced from field data. J Hydrol 529:1293–1301

Serio F, Miglietta PP, Lamastra L, Ficocelli S, Intini F, De Leo F, De Donno A (2018) Groundwater nitrate contamination and agricultural land use: a grey water footprint perspective in Southern Apulia Region (Italy). Sci Total Environ 645:1425–1431

Stuart ME, Gooddy DC, Bloomfield JP, Williams AT (2011) A review of the impact of climate change on future nitrate concentrations in groundwater of the UK. Sci Total Environ 409:2859–2873

USEPA (U.S. Environmental Protection Agency) (2004) Risk assessment guidance for superfund volume I: human health evaluation manual (part E, supplemental guidance for dermal risk assessment). Office of Superfund Remediation and Technology Innovation, Washington, DC

USEPA (U.S. Environmental Protection Agency) (2011) Exposure factors handbook: 2011 edition. United States Environmental Protection Agency, Office of Research and Development C., Washington, DC EPA/600/R-09/052F. http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf

Walraevens K, Mjemah IC, Mtoni Y, Van Camp M (2015) Sources of salinity and urban pollution in the Quaternary sand aquifers of Dar es Salaam, Tanzania. J Afr Earth Sci 102:149–165

Wen X, Feng Q, Lu J, Wu J, Wu M, Guo X (2018) Risk assessment and source identification of coastal groundwater nitrate in northern China using dual nitrate isotopes combined with Bayesian mixing model. Hum Ecol Risk Assess 24:1043–1057

Wen X, Lu J, Wu J, Lin Y, Luo Y (2019) Influence of coastal groundwater salinization on the distribution and risks of heavy metals. Sci Total Environ 652:267–277

Wu J, Lu J, Luo Y, Duan D, Zhang Z, Wen X, Min X, Guo X, Boman BJ (2016) An overview on the organic pollution around the Qinghai-Tibet plateau: the thought-provoking situation. Environ Int 97:264–272

Wu J, Zhao W, Lu J, Jin S, Wang J, Qian J (2018) Geographic information system based approach for the investigation of groundwater nitrogen pollution near a closed old landfill site in Beijing, China. Environ Eng Manag J 17:1095–1101

Xue D, Botte J, De Baets B, Accoe F, Nestler A, Taylor P, Van Cleemput O, Berglund M, Boeckx P (2009) Present limitations and future prospects of stable isotope methods for nitrate source identification in surface-and groundwater. Water Res 43:1159–1170

Yang L, Han J, Xue J, Zeng L, Shi J, Wu L, Jiang Y (2013) Nitrate source apportionment in a subtropical watershed using Bayesian model. Sci Total Environ 463–464:340–347

Yang J, Huang D, Zhang L, Xue W, Wei X, Qin J, Ou S, Wang J, Peng X, Zhang Z, Zou Y (2018) Multiple-life-stage probabilistic risk assessment for the exposure of Chinese population to PBDEs and risk managements. Sci Total Environ 643:1178–1190

Zhai Y, Zhao X, Teng Y, Li X, Zhang J, Wu J, Zuo R (2017) Groundwater nitrate pollution and human health risk assessment by using HHRA model in an agricultural area, NE China. Ecotox Environ Safe 137:130–142