
Nitrate pollution of groundwater in the Yellow River delta, China

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Abstract Nitrate pollution of groundwater in the Yellow River delta, China is an important issue related not only to nitrate dispersion and health concerns but also to mass transport and interactions of groundwater, sea, and river waters in the coastal area. The spatial distribution of nitrate, nitrate sources, and nitrogen transformation processes were investigated by field surveys and geochemical methods. Nitrate occurred mainly in shallow layers and had a spatial distribution coinciding with geomorphology and land/water use. Irrigation water from the Yellow River and anthropogenic waste are two main nitrogen sources of nitrate in the delta, and both denitrification and mixing processes could take place according to characteristics identified by ionic and isotopic data.

Résumé La pollution par les nitrates des eaux souterraines dans le delta de la rivière Jaune en Chine est un sujet important pas uniquement pour les problèmes de la dispersion des nitrates et de santé mais aussi du point de vue du transport de masse et des interactions entre les eaux souterraines, la mer et les eaux de rivière dans la zone côtière. La distribution spatiale des nitrates, des sources de nitrates et des processus de transformation des nitrates ont été étudiés grâce à des études de terrain et des méthodes géochimiques. Les nitrates sont principalement localisés dans les couches de surface et présentent une distribution spatiale qui coïncide avec la géomorphologie et les usages de l'eau et du sol. L'eau d'irrigation issue de la rivière Jaune et les déchets anthropiques constituent deux sources principales de nitrates dans le delta, et la dénitrification et les processus de mélange pourraient avoir lieu d'après les caractéristiques identifiées grâce aux données ioniques et isotopiques.

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Resumen La contaminación con nitratos de agua subterránea en el delta del Río Amarillo, China, es un problema importante relacionado no solo con la dispersión de nitratos y temas de salud sino también con el transporte de masa e interacciones de agua subterránea, mar, y aguas fluviales en el área costera. Se investigaron la distribución espacial de nitrato, fuentes de nitrato, y procesos de transformación de nitrógeno mediante levantamientos de campo y métodos geoquímicos. Los nitratos se presentaron principalmente en capas someras y tuvieron una distribución espacial que coincide con la geomorfología y el uso del agua y la tierra. Las dos fuentes principales de nitrato en el delta son el agua de riego del Río Amarillo y residuos antropogénicos, y de acuerdo a las características identificadas mediante datos iónicos e isotópicos, pueden ocurrir procesos tanto de mezcla como de desnitrificación.

Keywords Groundwater flow · Land and water use · Nitrate pollution · Stable isotopes · Yellow River

Introduction

The modern Yellow River delta, China formed between 37° 11'–38° 13'N and 118° 11'–119° 15'E after 1855 when dyke breaching occurred at Tongwaxiang in Henan Province (Yu 2002). The modern delta has a coastline of approximately

350 km, an alluvial fan shape, and an area of approximately 5,400 km² (He et al. 1999), extending from the apex at Ninghai to the Taoer River mouth in the north, and to the Zhimai River mouth to the southeast (Fig. 1). A new alluvial fan has been created within the modern delta with the apex at Yuwa and an area of approximately 2,200 km² (Gao et al. 1989). The highest elevation in the delta is approximately 13.3 m above mean sea level (amsl) in the west; the lowest elevation is 1 m along the coast. The average topographical gradient is approximately 1/8,000–1/2,000. Quaternary sediment is approximately 300–500 m deep in the delta with interfingering aquifers of either marine or terrestrial layers. The sandy or fine sandy layers of Holocene and late Pleistocene, serving as the aquifer, are located approximately at 9–12, 20–22, and 26–37 m depth; these layers are overlapped with silt and/or clay (Cheng et al. 1987; Gao et al. 1989).

The monsoon climate brings rainfall to the delta mainly in summer (June–September). Annual average rainfall is approximately 594 mm, while potential evaporation is approximately 2,049 mm. The sediment production of the whole basin is estimated to be 1.6×10^9 metric tons/year, and 1.2×10^9 tons are transported to the delta area, of which 80% is deposited in the delta and coastal zone and 20% is carried to the ocean. High sediment load contributes to the enlargement of the delta and upward rising of the riverbed, which is approximately 3–4 m higher than adjacent riparian areas and has increased about 2.26 m at Lijin since the 1950s (Ye et al. 1997). The delta area has increased approximately 20–25 km²/year, and the mouth of the Yellow River has extended approximately 30 km from 1976 to 1999 though erosion at the mouth of the old abandoned channel. With the increasing area and elevation of the delta, changes in water flow, mass

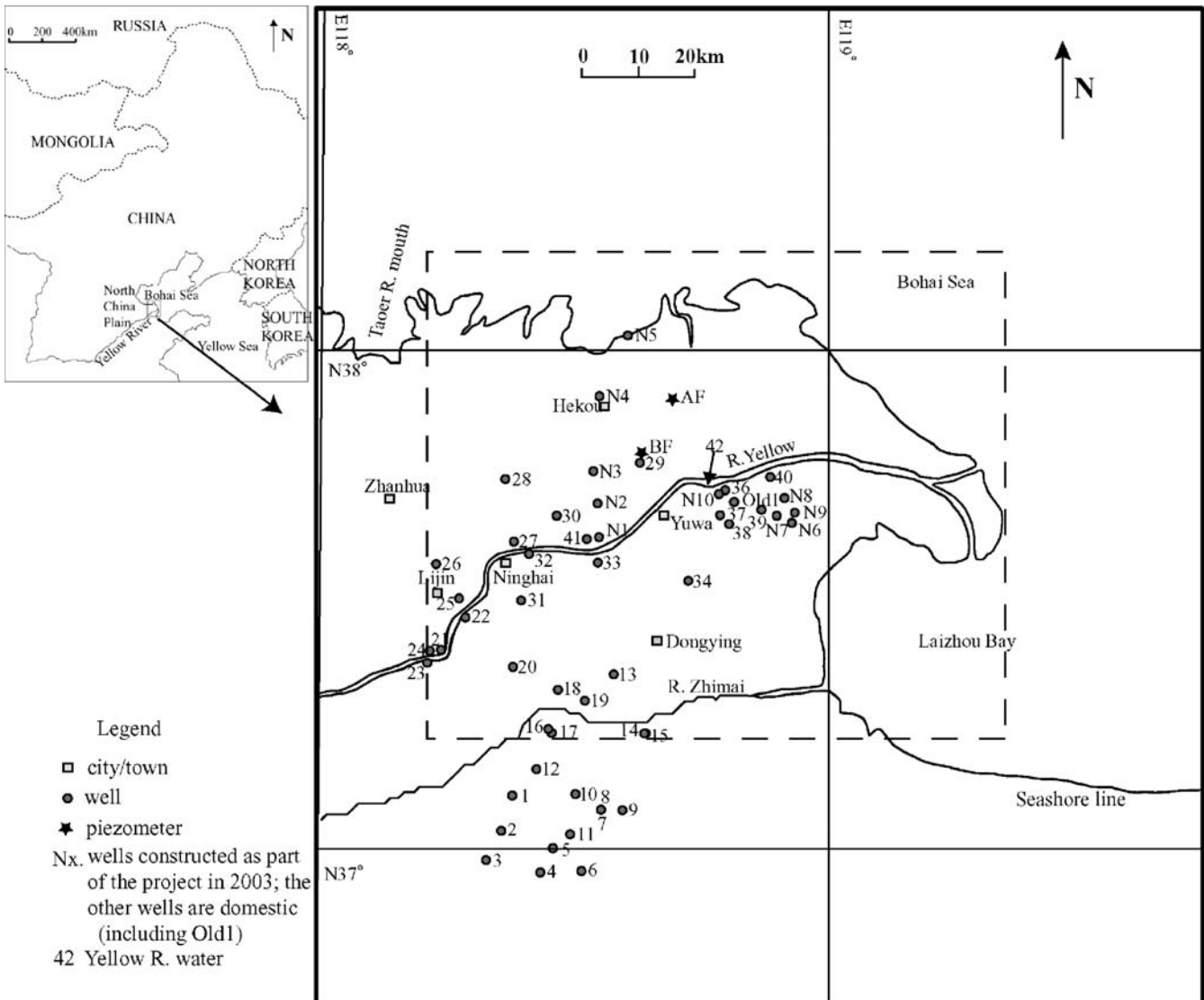


Fig. 1 Map of the study area and location of water sample sites in the Yellow River delta. The area within the *dashed line* relates to the geomorphologic map in Fig. 5

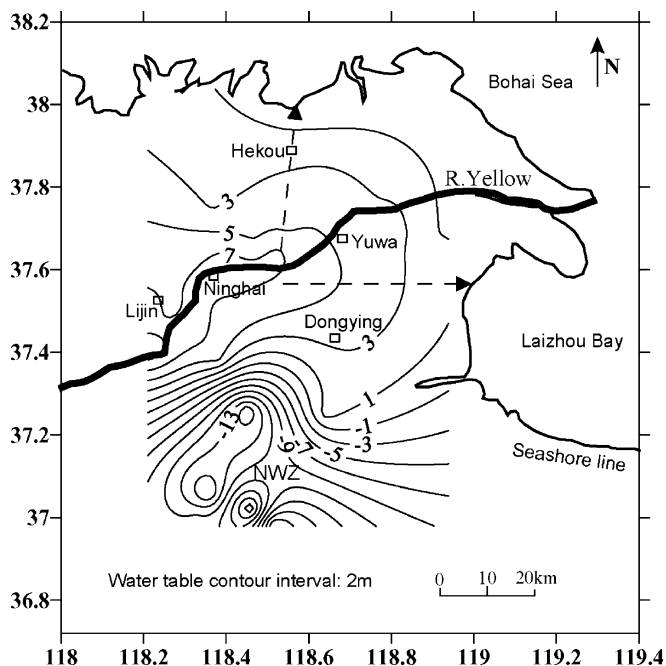


Fig. 2 Contour map of the groundwater table (unit: amsl) measured in Sept. 2003. Dashed lines with arrows indicate the direction of groundwater flow. NWZ is a zone where the water table is below mean sea level

transformation, and the interaction of sea, groundwater, and river waters can be anticipated.

Land reclamation for agriculture, by diverting irrigation water from the Yellow River, began in the 1950s; population in the area grew with the reclamation of more agricultural land and the later development of new oil fields. The area of arable land reached a maximum in 1984 and later decreased due mainly to urban development (Liu et al. 2003). The main crop in the delta is cotton with a limited area of winter wheat and paddy rice. Groundwater is unsuitable for irrigation due to high salinity. Twenty-one large reservoirs with capacities exceeding $1.0 \times 10^6 \text{ m}^3$ each were constructed to store water diverted from the Yellow River. The total capacity of all the reservoirs in the delta is approximately $4.85 \times 10^8 \text{ m}^3$. Although the nitrate level in the Yellow River is less than 45 mg/L (meeting the World Health Organization drinking water standard; Follett 1989) and the river water does not cause diseases such as methemoglobinemia when used as drinking water, nitrate pollution of the groundwater is an important issue in the delta for the following reasons.

The Yellow River delta is composed of large wetland areas, where denitrification may reduce nitrate into N gases in a reducing environment; i.e., the wetland may function to clean the earth through natural purification processes. This raises the question as to whether N-transformation processes in the groundwater occur in the delta.

Active interactions among sea, groundwater, and river waters take place in the delta, and large amounts of nutrients are transported from the continent to the ocean through either the surface channel or the aquifer. Generally, groundwater contains more nutrients than surface

water, and thus a high output of nutrients to the ocean through groundwater flow (e.g., submarine groundwater discharge, SGD) is expected (Taniguchi et al. 2002).

As saline groundwater is replaced by freshwater in the lower reaches of the river and in the delta area through irrigation and seepage from the Yellow River, the boundary between saline and freshwater moves eastward. Groundwater is thus possibly becoming fresher and may be a potential water supply resource in the delta.

This study sought to identify the spatial distribution, sources, and N processes of nitrate in the groundwater and to examine the association of nitrate with land and water use in the Yellow River delta based on field surveys, piezometer measurements, and chemical and isotopic (^{18}O , ^2H or D, ^{15}N) features.

Materials and methods

Field surveys were carried out in August 2002, September 2003, May and September 2004, and May 2005, and 52 water samples were collected in 2003 to examine nitrate contents (Fig. 1). Most of the groundwater samples were collected from wells inside or close to residential areas; these wells were used mainly for washing, except those in the southern part of the study area, which are used for potable supply. The majority of the wells were less than 20 m in depth. Ten new wells were constructed and labeled N1, N2 ... N10, as shown in Fig. 1, to investigate two groundwater flow routes from the Yellow River to the north and to the east. The ten wells were 20 m deep with two screens, which varied from well to well at depths of 2–6 m for the upper screens and 10–19 m for lower screens. A water sampler that could take water samples at

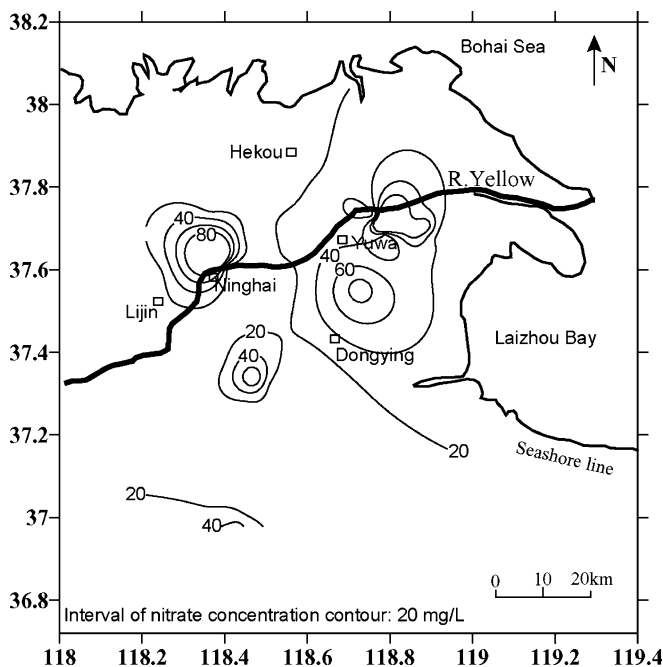


Fig. 3 Nitrate concentration contours of groundwater sampled in Sept. 2003

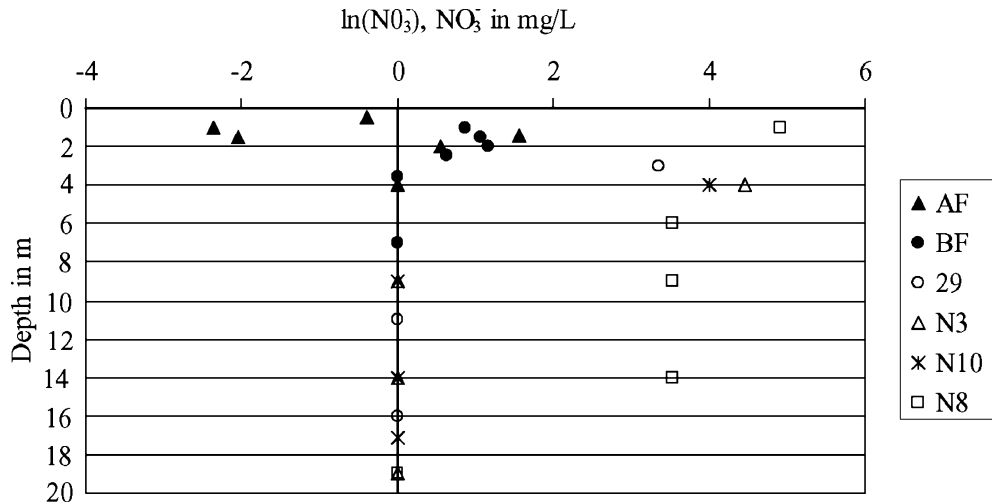


Fig. 4 Vertical profiles of nitrate concentration in four wells (Nos. 29, N3, N8, N10) in a residential area and two sites AF and BF from piezometers and porous cup samplers in a cropped area. Indicated $\ln(NO_3^- = 0)$ as 0 in the y-axis

given depths was used to investigate the vertical change in nitrate concentration during May 2005. Because few wells existed in the northeast part of the study area, two groups of piezometers were placed into a crop field in May 2005 at two depths: 1.42 and 4.0 m at site AF and 3.56 and 6.98 m at site BF (Fig. 1). These two locations were far from residential areas. Porous suction cups were set up to collect soil water at four depths: 0.5, 1, 1.5, and 2 m at AF and 1, 1.5, 2, and 2.5 m at BF.

Water samples were analyzed for NO_3^- , Cl^- , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} , and Ca^{2+} by ion chromatography and HCO_3^- by titration (pH 4.8 alkalinity). The spectrophotometric method with phenol disulfonic acid was used to analyze NO_3^- when salinity was too high for NO_3^- to be detected in a diluted sample. The stable isotopes ^{18}O , D, and ^{15}N were analyzed using a Delta S mass spectrometer in units of standard mean ocean water (SMOW) for ^{18}O , D, and AIR (referred to atmospheric N_2 standard) for ^{15}N . ^{15}N was analyzed with the following procedure: absorption of NO_3^- in anion resin rinsed later by 2N HCL, transformation of NO_3^- to NH_4Cl by adding NaOH and Devarda's alloy, and speciation and purification of N_2 gas (Mitsui et al. 1980). Scintillation detectors were used to measure ^{14}C for preprocessing water samples in the field, i.e., obtaining $BaCO_3$ sediment by adding NaOH and $BaCl_2$ (Chen et al. 2003).

Dissolved oxygen (DO), electrical conductivity (EC), pH, oxygen reduction potential (ORP), water level, and well depth were measured in situ. Well elevations were obtained from a 1:10,000-scale map, and their locations plotted using a geographical information system (GIS). Information on groundwater use, screen depth, and land use was obtained through communications with well managers and farmers.

Results

Groundwater flow

The channel of the Yellow River serves as a boundary between surface and groundwater flow in the delta;

groundwater flows northward in the northern part and eastward in the southern part of the study area. Water table contours measured in September 2003 show the flow directions (Fig. 2). Potential gradients of groundwater flow in the northward and eastward directions based on the contour lines in Fig. 2 were estimated to be 1/4,500–1/4,000 and 1/7,000, respectively. Pumping tests on the ten new wells showed transmissivity of 0.036–0.61 m^2/day . A zone where the water table is below mean sea level (NWZ) was found in the far southern part of the study area (Fig. 2). Groundwater in the NWZ, flowing from the upper lands of the alluvial delta fan, has low EC, and has been overpumped for irrigation and domestic water uses for the last 20–30 years.

In addition to the NWZ, for approximately 226 days in 1997, no water flowed in the delta river channel, and the water table dropped dramatically to a level lower than 0 m amsl in an area east of Yuwa. The water table in this area in 1997 was approximately 5 m lower than in 1991 (He et al. 1999), indicating that the zone was affected greatly by Yellow River water.

Range of nitrogen concentration

Among 52 water samples collected in 2003, 49 had nitrate (as NO_3^-) concentrations greater than 1 mg/L; the average nitrate concentration for all samples was about 30 mg/L with a maximum of 202 mg/L. Although nitrate is rather stable in groundwater, it changes seasonally to some extent. The nitrate concentration was 62 and 64 mg/L at wells 38 and 39, respectively, in 2002 and changed to 53.8 and 101 mg/L, respectively, in 2003. The nitrate concentration of Yellow River water was measured as 13.6, 19.5, and 20.6 mg/L in August 2003, September 2003, and May 2005, respectively. Because approximately $1.2 \times 10^9 m^3$ water per year is actually diverted from the Yellow River delta for irrigation, industrial, and domestic uses (He et al. 1999), nitrate in the river water poses a health threat and may contribute to the pollution of groundwater. The percentage of water used for irrigation to the total water

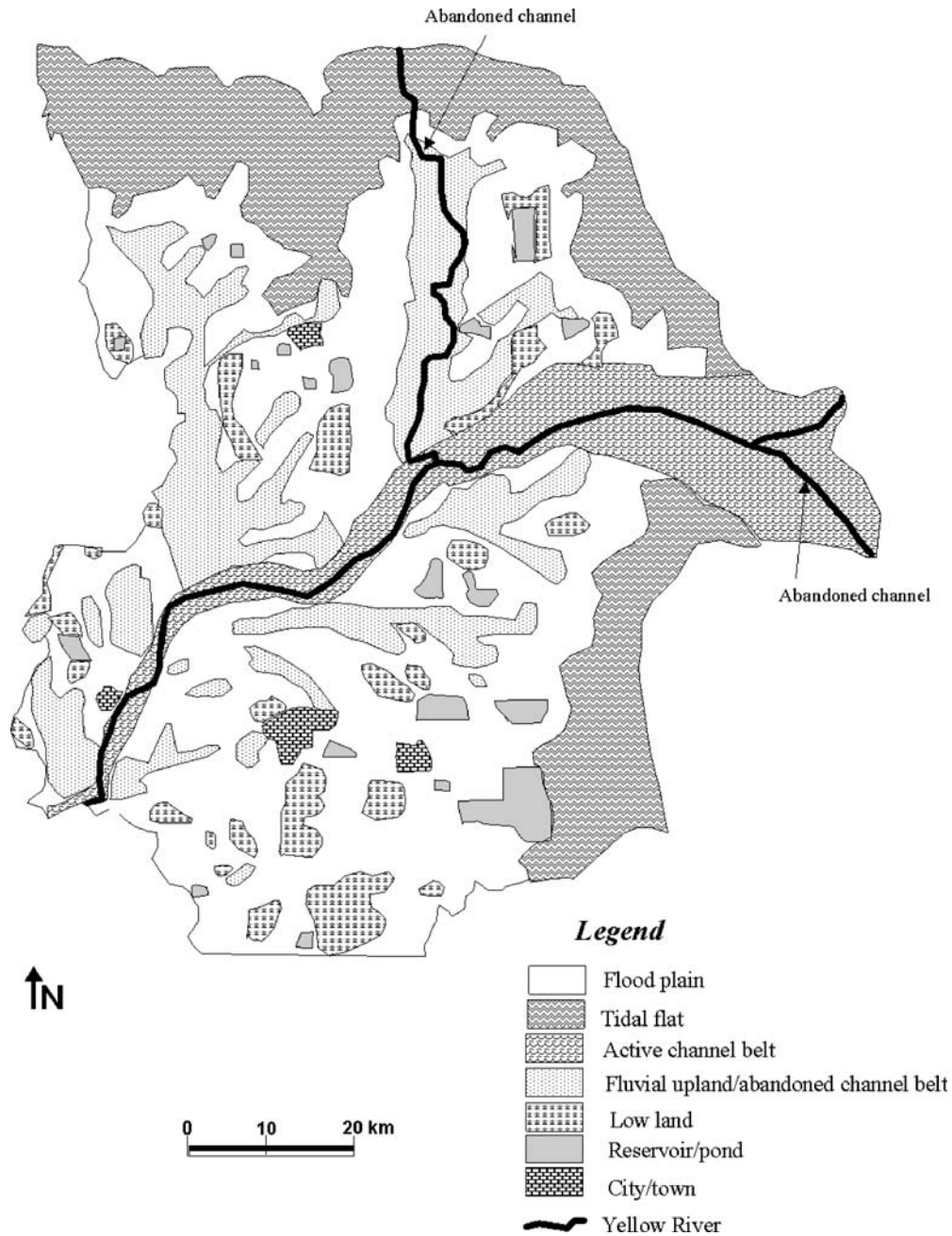


Fig. 5 Geomorphology of the Yellow River delta (modified from He et al. 1999; Liu et al. 2004)

diverted from the Yellow River is approximately 50%. Thus, given the average nitrate concentration of 17.9 mg/L in the river water, about 1.07×10^7 kg of NO_3^- pours annually into agricultural fields.

Spatial distribution of nitrate in groundwater

Figure 3 shows the horizontal distribution of nitrate in groundwater with two zones of elevated concentration close to Ninghai and Yuwa along the Yellow River and another zone about 20 km south of the river. The distance from the coastline to the center of the zone near Yuwa is only about 20 km, indicating a threat to the coastal ecological system.

This zone roughly corresponds to the area that experienced a noticeable drop in the water table in 1997 and has a water table of approximately 3–5 m amsl, as indicated in Fig. 2. The zone near Ninghai is close to the northern part of the river and has a relatively high water table of 3 to 7 m amsl. The third zone of elevated nitrate concentration is relatively small and located near the western district and Dongying City, where the aquifer has two layers, with brackish water in the shallow layer and freshwater in the deep layer (He et al. 1999).

Because the well screens in the delta had water from different layers, i.e., mixed water, in 2003, it was hard to judge the nitrate depth using the data from the water

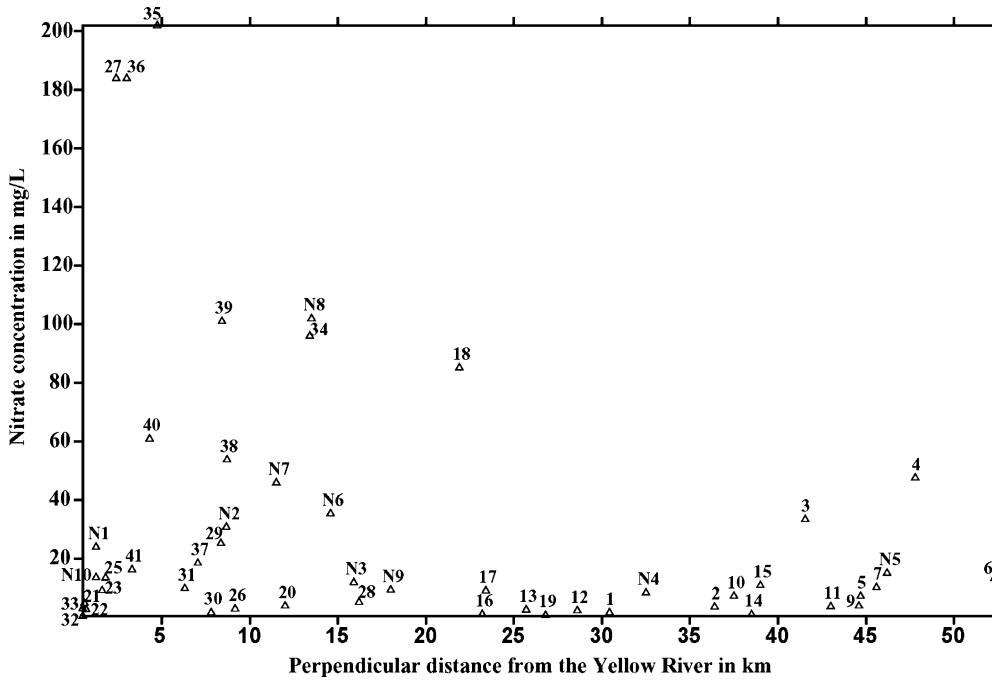


Fig. 6 Change in nitrate concentration along a route perpendicular to the Yellow River

samples. Figure 4 shows the vertical profile of nitrate concentrations in the wells from which groundwater at varying depths was collected in 2005 using a new sampler. Nitrate was found only in shallow layers less than 4 m deep, except for well N8, indicating the mixing of water from the different layers. All four wells were located in a residential area close to a manure pit, resulting in the high-nitrate content. Sites AF and BF were selected to investigate the vertical profile of nitrate (Fig. 4). A nitrate concentration of less than 5 mg/L, much less than that in the residential area, was detected only for samples collected in the shallow layer less than 3 m deep. In the layer deeper than 3 m, no nitrate was detected, as in the wells from the residential area.

6–8 m and 12–15 m and a peat layer 50–60 cm thick at approximately 15 m in depth. The water table of the deep well (30 m depth) was 27.5 cm higher than that of the shallow well (10 m depth) at the core site on 5 November 2005, indicating a characteristic of upward potential in the discharge area. On the other hand, the water tables in the shallow layer shown by the two piezometers at AF were 0.41 m (piezometer depth: 1.42 m) and 1.49 m (piezometer depth: 4.0 m) deep, and those at BF were 1.78 m (piezometer depth: 3.56 m) and 3.43 m (piezometer depth: 6.98 m) deep in May 2005, indicating a downward potential gradient, in contrast to the deep layer. The reasons for the downward gradient in the shallow zone could be the existence of the structure of multiple layers and/or the perched layer. Pressure sensors set up in the piezometers indicated water-table fluctuations responding

Discussion

Controls on the current spatial distribution of nitrate

Chen et al. (2005) found that the spatial distribution of nitrate closely relates to the groundwater flow system in the North China Plain. In the discharge area near the Yellow River, nitrate occurred in shallow layers of less than 8 m; the upward potential was considered the main reason for this distribution. The stratum in the delta, which is composed of integrated layers of marine and terrestrial depositions, is much more complex than that in the North China Plain. Gao et al. (1989) reported that seawater intrusion has occurred three times in the delta in the last 10,000 years. A bore core to a depth of 30 m collected in the town of Liuhu, about 10 km southeast of Dongying (western district), showed two layers of shells at depths of

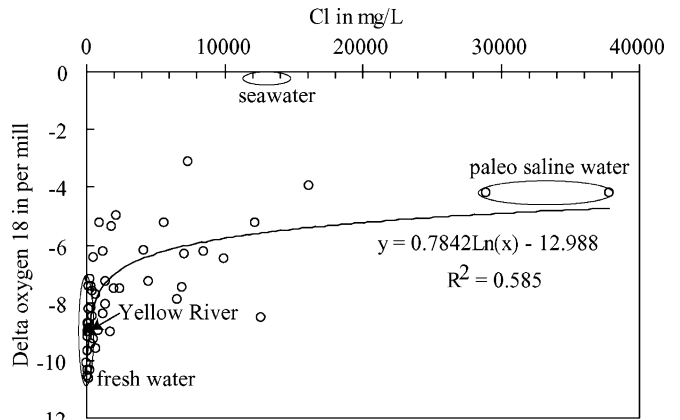


Fig. 7 Differentiation of water sources using chloride concentration and $\delta^{18}O$ values. The curved line represents the mixing process

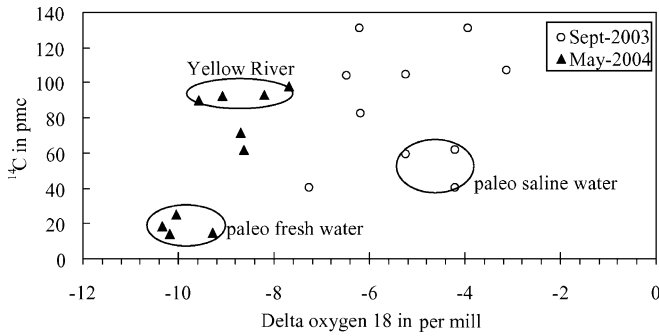


Fig. 8 Identification of water sources by $\delta^{18}\text{O}$ and ^{14}C values in water sampled in May 2004 and Sept. 2003; *non-circled points* showed a mixing with varied sources

to rainfall within a range of about 1 m, enabling a mixture of chemical components within a layer. Within the regional groundwater flow system (Toth 1999), the delta area is regarded as the discharge zone, and upward gradient of the deep layers is the “obstacle”, which keeps the nitrate in the upper zone.

Sediment depositions and shifts of the Yellow River contribute predominantly to the geomorphological formation in the delta, where abandoned channels are generally higher than the riparian area, while the area sandwiched between the abandoned channels becomes the lowland. Two

pieces of upland (the abandoned channels) with a relatively coarse deposition were created in the northern part of the Yellow River delta from 1904 to 1926 (western piece) and 1953–1976 (eastern piece; Fig. 5). Originally, these uplands all had better groundwater quality and resources than the lowlands; upland areas were reclaimed first for agricultural uses and new towns and/or residential areas. Two large irrigation projects in the Yellow River delta irrigated a total area of $5.2 \times 10^2 \text{ km}^2$ (CDCID 2002). Given this situation, the correspondence between the two zones of high-nitrate content shown in Fig. 3 and the two pieces of upper land is not surprising. A third concentration zone is located near Dongying City, which has well developed agriculture and a high density of residential housing.

The impact of the Yellow River on nitrate pollution of groundwater was examined further by the relationship between nitrate concentration and distance perpendicular to the Yellow River (Fig. 6). Nitrate concentration generally decreased as the distance increased, and 20 km is regarded as the approximate border of the impact zone with the Yellow River in terms of nitrate pollution. Previous research has shown that the Yellow River impacts groundwater flow in the lower reaches of the river and the delta in a zone approximately 20–40 km from the river, based on water level changes, chemical components, and isotopic data (Chen et al. 2001, 2005).

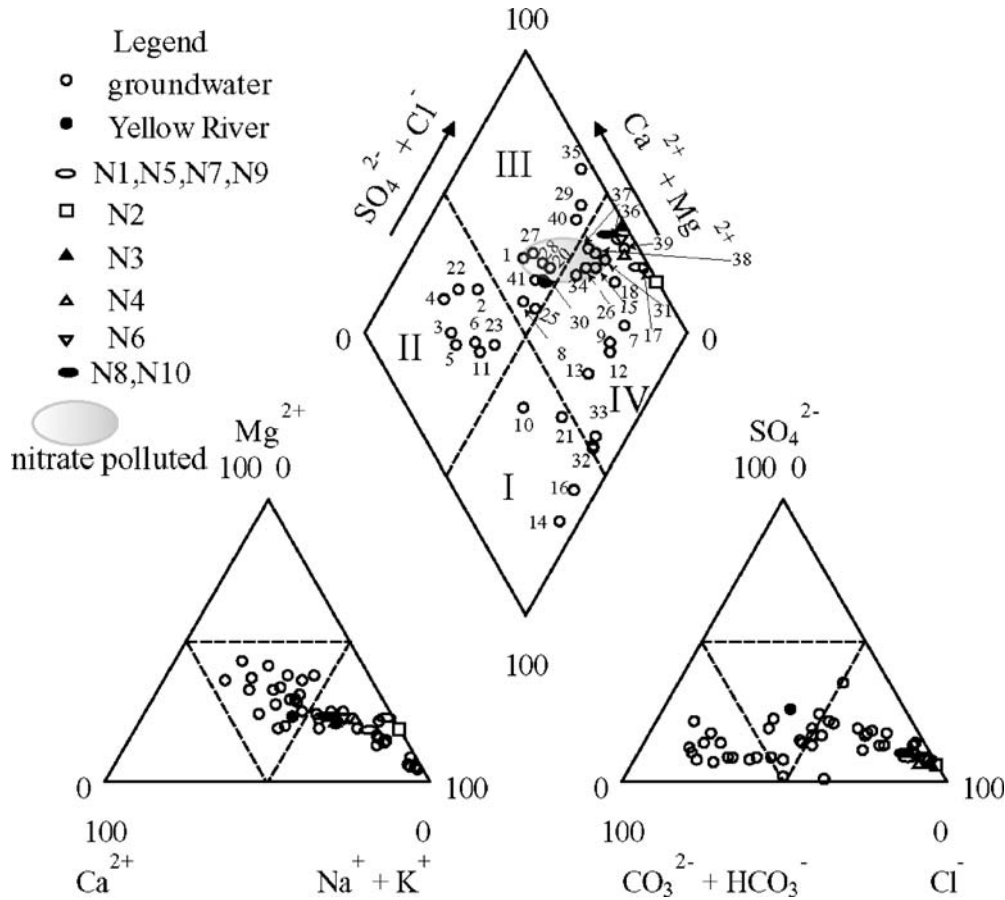


Fig. 9 Piper diagram for the major ions in the Yellow River delta; nitrate polluted samples are indicated by the ellipse

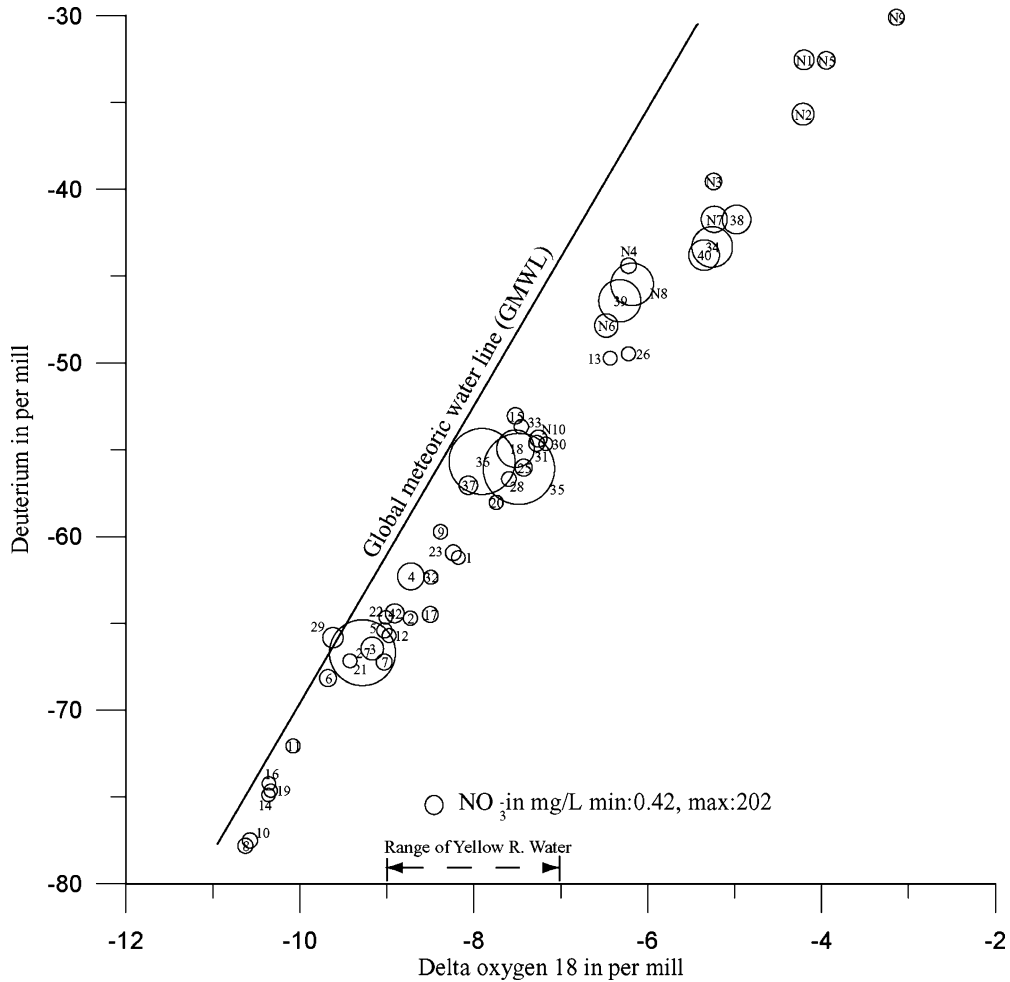


Fig. 10 Cross plot of $\delta^{18}\text{O}$ and δD with nitrate concentration indicated by proportionate circles

Areas farther than 40 km from the river have high-nitrate concentrations, indicating a different water source.

Sources of nitrate pollution in groundwater

Four water sources affect the groundwater in the delta: Yellow River water, paleo-freshwater, paleo saline water, and seawater. The first two sources can be grouped as freshwater because both are low in chloride concentration (Fig. 7). The groundwater affected mainly by the Yellow River had a high concentration of ^{14}C in percent modern carbon (pmc), indicating an age of less than 50 years (Fig. 8). Paleo fresh groundwater was dated by ^{14}C to between 11,000 and 15,800 BP (corresponding to 25.3 and 14.7 pmc respectively), while paleo saline water was dated to between 4,000 and 7,400 BP (corresponding to 61.5 and 40.6 pmc respectively) (Fig. 8). Generally, both Yellow River water and paleo freshwater had low chloride at less than about 300 mg/L. Values of $\delta^{18}\text{O}$ for river water were -9 to -7‰ , while those for paleo-freshwater were less than -9‰ . For seawater, $\delta^{18}\text{O}$ was close to 0‰ , and chloride content was 11,000–15,000 mg/L, which is equivalent to an EC value of approximately 30–40 mS/cm. Paleo saline water had a very high chloride value of 30,000 to 40,000

mg/L and $\delta^{18}\text{O}$ value of approximately -4 to -3‰ . Figure 7 shows an interaction curve with a logarithm shape, indicating the mixing of all these sources.

The Piper diagram in Fig. 9 shows the chemical component of the water samples grouped into four classes. Classes I and II, located mainly in the far southern part of

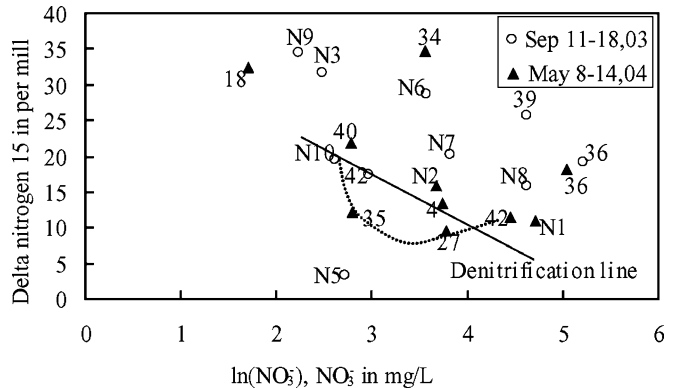


Fig. 11 Denitrification (straight line) and mixing processes (dashed line) inferred from the relationship between $\delta^{15}\text{N}$ and $\ln(\text{NO}_3)$. No. 42 is Yellow River water. Data given for 11–18 September 2003 and 8–14 May 2004

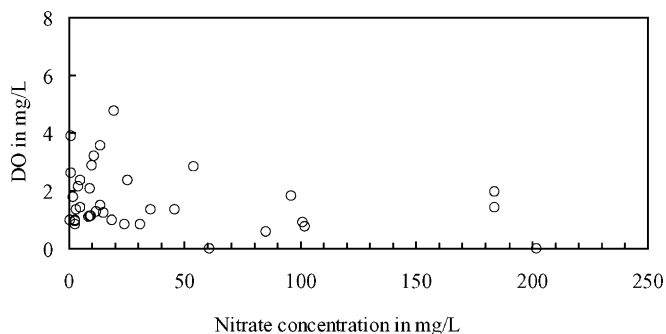


Fig. 12 Cross plot of nitrate concentration and DO showing that the majority of the samples, including those with high nitrate, have low DO

the study area, had relatively low contents of SO_4^{2-} and Cl^- . Class III had high contents of Ca^{2+} and Mg^{2+} and was influenced greatly by the Yellow River. Class IV had high contents of Na^+ and K^+ and was affected by either seawater or paleo saline water. Nitrate pollution water samples chiefly related to classes III and IV, as shown in the figure.

Figure 10 shows the relationship between stable isotopes, $\delta^{18}O$ and δD , and nitrate indicated by circles, the radii of which linearly relate to the nitrate concentration. The correlation between nitrate pollution and the Yellow River was observed, as in Fig. 9, because high-nitrate concentration corresponded to $\delta^{18}O$ values of -9 and -7% , which fall in the range of Yellow River water.

$\delta^{15}N$ was found at 10 – 22% in manure/urine and 2 – 9% in natural soil organic-N (Kreitler and Jones 1975; Heaton 1986). Because Yellow River water showed $\delta^{15}N$ values of 11.53% (May 2004) and 17.45% (Sept. 2003; Fig. 11), nitrate in the river water is assumed to have derived from anthropogenic waste. Therefore, irrigation using river water and the use of waste as organic fertilizer could be two sources for those samples with a $\delta^{18}O$ value of -9 to -7% and a $\delta^{15}N$ of 10 – 22% . Manure pits without boundary linings inside residential areas are likely sources for samples with $\delta^{15}N$ values of 10 – 22% , but $\delta^{18}O$ higher than -7% . Nitrate at N5, located in a fish and shrimp cultivation area, may have derived from inorganic fertilizer.

Nitrogen transformation processes implicated

Nitrogen from natural (rainfall and geologic formations) and the other sources (forage and pastoral agriculture, fertilizers, and waste disposal) undergoes mineralization, immobilization, fixation, nitrification, and denitrification (Komor and Anderson 1993). These transformation processes are related closely to water movement in the continuum of groundwater, soil, plant, and atmosphere. Because the delta is basically a reducing environment, denitrification is expected to reduce nitrate to nitrogen gases, which then return to the atmosphere.

As an essential condition for denitrification, DO must usually be at a low level of less than 2 mg/L. Most of the DO measured in situ was lower than 2 mg/L, but some of these low DO samples had high-nitrate concentration

(Fig. 12). Although both denitrification and the mixing process gave similar curves for the relationship between nitrate and $\delta^{15}N$, Mariotti et al. (1988) verified that the denitrification process, which follows the rule of Rayleigh fractionation, results in a linear relationship between the log of nitrate concentration and $\delta^{15}N$. The observed relationship in this study indicates that both denitrification and mixing processes may occur at wells 4, 27, 35, 42, 40 and N10 (Fig. 11), which have a common nitrate source. The straight line in Fig. 11 shows the denitrification process, while the dashed curve shows the mixing process. It is still hard to judge the processes for the other sources of water and nitrate.

Conclusions

The zones of elevated nitrate concentration corresponded with the distribution of fluvial upper land formed mainly by abandoned channels and reclaimed for agriculture by using irrigation water diverted from the Yellow River. Multiple layers of aquifer and upward potential gradients prevent nitrate from moving lower into the deep layers. Thus, nitrate was detected only in the shallow layer with depths less than 3 – 4 m. The Yellow River provides water for irrigation and nutrients for crops and has been identified as an important source of nitrate in groundwater with $\delta^{15}N$ values ranging from 11.5 to 17.5% and $\delta^{18}O$ values from -9 to -7% . A nitrate impact zone reaching approximately 20 km from the Yellow River was found by examining the relationship between nitrate concentration and distance perpendicular from the river. Anthropogenic sources such as manure pits, are another source of nitrate pollution in the delta and account for $\delta^{15}N$ values of 10 to 20% . Both denitrification and the mixing process could occur within the nitrogen cycle, and contribute to the observed distribution of nitrate.

Although groundwater is not commonly used as drinking water in the modern delta, nitrate in groundwater remains a critical issue associated with mass transport, waterway transformation, and interactions of surface, ground, and seawaters along the coastal zone.

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