Source Apportionment and Transport-Transformation Patterns of Nitrate in the Fuhe River-Baiyangdian Lake System (Post-print)

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Abstract

In recent years, rapid economic development and population growth in the Baiyangdian watershed have led to increased domestic sewage discharge, seriously threatening the water quality of the Fu River and Baiyangdian. Among these issues, eutrophication caused by excessive nitrate concentrations represents a significant challenge facing river systems. Taking Baiyangdian and its only perennial inflow river—the Fu River—as the study area, this research employs hydrochemistry, hydrogen and oxygen isotopes in water ($ $2H, $ 18*O*), *andnitratenitrogenisotopes*($15N) to analyze the hydrochemical characteristics and changes in water chemistry types from 2008 to 2016, identify the sources of nitrate pollution and its migration and transformation patterns along the Fu River-Baiyangdian area, and provide references for eutrophication management. The results indicate that: in 2008, the nitrate $ $15N values in the Fu River were >10‰, and in 2014, the nitrate $ $15N values ranged from 2.07‰ to 18.49‰, indicating that nitrate in the Fu River primarily originated from domestic sewage from Baoding City and villages along the river; however, in 2009, the nitrate $ $15N values ranged from -3.7‰ to 4‰, suggesting that nitrate mainly came from industrial wastewater. In the Baiyangdian lake area, the nitrate $ $15N values ranged from 5.8‰ to 11.7‰ in 2008 and from 3.31‰ to 12.53‰ in 2014, while in 2009, the $ $15N values ranged from -3.8‰ to 0.7‰, demonstrating that domestic sewage and industrial wastewater from the Fu River constitute the primary sources of nitrate in the Baiyangdian lake area. From 2008 to 2014, the concentration ratios of Cl and SO4 2 gradually decreased, indicating that the discharge of industrial wastewater and domestic sewage was being controlled; in 2009, NO3 concentrations exceeded 50 mg · L 1 due to industrial wastewater discharge, while in 2014 and 2016, NO3 concentrations were within standards; the primary factors controlling nitrate concentration variations were precipitation dilution, external inputs, and denitrification. When dissolved oxygen (DO) was less than 2 mg·L 1, nitrate reduction was primarily influenced by denitrification.

Full Text

Sources, Migration and Transformation of Nitrate in the Fuhe River and Baiyangdian Lake, China

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Abstract

Baiyangdian Lake, the largest freshwater lake in the North China Plain, plays a critical role in flood control, microclimate regulation, ecological improvement, and the development of aquaculture and tourism. However, rapid economic development and population growth in the watershed have led to sharply increased pollutant loads, with nitrate-induced eutrophication representing a major challenge for the aquatic system. This study investigated the Fuhe River—the only perennial inflow tributary to Baiyangdian—and the lake itself, using hydrochemical analysis combined with hydrogen and oxygen isotopes (δ^2H, δ^{18}O) and nitrate nitrogen isotopes (δ^{15}N) to characterize water chemistry, identify nitrate pollution sources, and elucidate migration and transformation processes from 2008 to 2016.

The results demonstrate that nitrate in the Fuhe River originated primarily from domestic sewage discharged from Baoding City and villages along the river channel, as evidenced by δ^{15}N values >10‰ in 2008 and ranging from 2.07‰ to 18.49‰ in 2014. In contrast, June 2009 showed δ^{15}N values between -3.7‰ and 4‰, indicating industrial wastewater as the dominant nitrate source. In Baiyangdian Lake, δ^{15}N values ranged from 5.8‰ to 11.7‰ in 2008 and 3.31‰ to 12.53‰ in 2014, while 2009 values of -3.8‰ to 0.7‰ confirmed that both domestic and industrial wastewater from the Fuhe River constituted the primary nitrate sources.

Between 2008 and 2014, the proportions of Cl^- and SO_4^{2-} gradually decreased, suggesting improved control over industrial and domestic discharges. While nitrate concentrations exceeded 50 $\mathrm{mg \cdot L^{-1}}$ in 2009 due to industrial wastewater input, levels remained within standards in 2014 and 2016. The primary factors controlling nitrate concentration variations were precipitation-driven dilution, external inputs, and denitrification. When dissolved oxygen (DO) fell below 2 $\mathrm{mg \cdot L^{-1}}$, denitrification became the dominant process responsible for nitrate reduction. These findings provide a scientific basis for eutrophication management in the Fuhe River-Baiyangdian Lake system.

Keywords: Fuhe River-Baiyangdian Lake Basin; Water quality; Eutrophication; Nitrate; Hydrogen and oxygen isotopes; Nitrogen isotope; Hydrochemical characteristics

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Introduction

Baiyangdian Lake, the largest freshwater lake in the North China Plain, serves vital functions in flood detention, climate regulation, ecological improvement, and regional aquaculture and tourism development [1]. However, rapid economic growth and population expansion in the watershed have caused pollutant loads to increase dramatically [2], with elevated nitrate concentrations triggering eutrophication that poses a serious challenge to the river system. The Fuhe River, the only perennial tributary among Baiyangdian’ s eight inflow channels, receives approximately 1\$×10^{5}\$ m^3 of domestic sewage and treated industrial wastewater daily from Baoding City, accounting for 45.24% of the river’ s average flow [7]. This unconventional water source accelerates eutrophication in Baiyangdian. Among various nitrogen forms causing eutrophication, the sources and biogeochemical transformation of nitrate (NO_3^-) in natural waters represent key research challenges. Excessive nitrate not only drives eutrophication but also threatens human health. In shallow riverbeds with high surface-area-to-volume ratios, nitrogen inputs undergo active biogeochemical transformations including nitrification, denitrification, and biological uptake, all controlled by external inputs and internal processes [3-4]. Therefore, identifying nitrate sources and understanding nitrogen cycling mechanisms are essential for effective water quality management.

Numerous studies have assessed surface water pollution from non-point sources in the Fuhe River-Baiyangdian system [5-7]. Since the 1970s, researchers have used nitrate nitrogen isotopes ($\delta^{15}N$) to trace nitrate origins, exploiting differences in isotopic composition among various nitrate sources and distinct fractionation mechanisms during nitrogen transformations. Segal-Rozenhaimer et al. [8] identified sewage discharge as the primary source of ammonium in the Jordan River using nitrogen isotopes to analyze nitrification processes along the river course. Wang et al. [3] found that increasing NO_3^--N and NH_4^+-N concentrations coupled with decreasing $\delta^{15}N$ values in the Wuchuan headwater stream resulted from combined effects of surface runoff nitrogen input and sediment-water interface nitrification. Wang et al. [9] applied $\delta^{15}N$ tracing to identify nitrogen pollutant sources in the Fuhe River, concluding that contaminants originated mainly from Baoding’ s industrial and domestic wastewater, with minimal contribution from agricultural non-point sources. However, spatiotemporal migration and transformation patterns of nitrate nitrogen along the Fuhe River-Baiyangdian continuum remain poorly understood. This study integrates stable nitrogen isotopes with hydrochemical analysis to investigate interannual variations in nitrate sources, concentrations, and transformation mechanisms in the Fuhe River surface water system, providing crucial insights for Baiyangdian water quality protection.

This research focuses on the Fuhe River and Baiyangdian Lake in the North China Plain with four objectives: (1) characterize interannual variations in major ions and water chemistry types in both water bodies from 2008 to 2016; (2) analyze seasonal and longitudinal patterns of hydrogen and oxygen isotopes; (3) identify nitrate pollution sources and assess contamination levels using δ^{15}N and NO_3^- concentrations; and (4) examine relationships between nitrate and hydrological parameters and other ions to elucidate migration and transformation processes in different years.

1.1 Study Area Description

The study area is located in the Baiyangdian watershed (38°43 -39°02 N, 115°45 -116°07 E) in central North China Plain, covering 362.8 km^2 [10]. The region experiences a temperate continental climate with mean annual precipitation of 510 mm, mean annual evaporation of 1,369 mm, and mean annual temperature of 13.8 °C. Precipitation is highly uneven, with 75% occurring during June–September, while 54% of evaporation concentrates in May–August [11-12]. The Fuhe River, a tributary of the Daqing River’ s southern branch, is the only perennial inflow river to Baiyangdian, receiving approximately 1$×10^{5}$ m^3 of domestic sewage and wastewater daily, which constitutes 45.24% of the river’ s average discharge [7].

1.2 Sampling and Analytical Methods

Field surveys and water sampling were conducted along the Fuhe River and Baiyangdian Lake. Eight sampling sites were established along the Fuhe River: upstream sites at Yindingzhuang (F1), Xianrenqiao Village (F2), and Nansun Village (F3); midstream sites at Xiaowangting Village (F4), Nanliukou Village (F5), and Shanmamiao Village (F6); and downstream sites at Anzhou (F7) and the Fuhe River Bridge at Anxin County (F8). Nine sampling sites were set up in Baiyangdian Lake. Sampling campaigns were conducted in September 2008, June 2009, June 2011, July 2014, and June 2016, with samples collected from upstream to downstream as shown in [Figure 1: see original paper]. Site selection considered sewage outfalls, topographic distribution, and flow distance to ensure representative coverage of typical regions along the Fuhe River-Baiyangdian continuum.

A total of 66 river and lake water samples were collected. Field measurements included temperature (T), electrical conductivity (EC), pH, dissolved oxygen (DO), and oxidation-reduction potential (ORP) using portable Horiba ES-71 (EC, T) and Horiba D-75 (pH, DO, ORP) meters. Samples were stored in 100 mL plastic bottles at 4 °C with minimal air contact after rinsing bottles twice with sample water. Laboratory analyses were completed within one week at the Key Laboratory of Water-saving Agriculture, Chinese Academy of Sciences. Bicarbonate (HCO_3^-) was determined by dilute sulfuric acid-methyl orange titration. Anions (F^-, Cl^-, SO_4^{2-}) and cations (Na^+, K^+, Mg^{2+}, Ca^{2+}) were measured by ion chromatography (ICS-2100, Dionex, USA) with acceptable ion balance errors within 5% [13]. Total dissolved solids (TDS) were calculated as the sum of all ions minus half of the HCO_3^- concentration [14].

Hydrogen and oxygen isotopes (δ^2H, $\delta^{18}O$) were analyzed using a liquid water isotope analyzer (L2120-i Isotopic H_2O; Picarro, USA). Nitrate nitrogen isotopes ($\delta^{15}N$) were enriched using a resin adsorption-elution method: (1) Anion exchange columns were prepared by adding 1.5 g chloride adsorption resin to syringes, rinsing with 10 mL of 3 $mol \cdot L^{-1}$ HCl, washing with 15 mL ultrapure water to remove excess chloride, and maintaining moisture with 0.5 mL ultrapure water before sealed refrigeration; (2) Adsorption: After field collection, 2 L water samples were filtered through 0.45 m membranes to remove particulates, then passed through columns at 500–1,000 $mL \cdot h^{-1}$ and transported to the laboratory with columns kept moist; (3) Elution: Nitrate adsorbed on columns was eluted with 3 $mol \cdot L^{-1}$ HCl, converted to silver nitrate by adding silver oxide, and freeze-dried to powder for $\delta^{15}N$ determination by stable isotope mass spectrometry (Isoprime100; Elementar, Germany) [15].

Nitrate concentration variations in surface water are influenced by dilution, external inputs, and biochemical reactions. Chloride (Cl^-) serves as an ideal conservative tracer due to its strong hydrophilicity and chemical stability. Based on chloride mass balance, measured NO_3^- concentrations were compared with calculated values to identify dominant controlling processes [16]: (1) If measured NO_3^- calculated NO_3^-, dilution and mixing control NO_3^- variation; (2) If measured NO_3^- > calculated NO_3^-, additional external NO_3^- inputs occur; (3) If measured NO_3^- < calculated NO_3^-, biochemical reactions such as denitrification affect NO_3^- concentrations. Calculated NO_3^- concentrations at sampling point *i* were determined using Equation (1), where NO and CO represent NO_3^- and Cl^- concentrations at the first sampling point (F1), and Ci and Ni denote Cl^- and calculated NO_3^- concentrations at point *i*, respectively. Cao et al. [16] applied this method to demonstrate that NO_3^- variation in the Songhua-Heilong River system was dominated by dilution, whereas excess nitrate input controlled NO_3^- changes in the Ussuri River.

2.1 Hydrochemical Characteristics of Fuhe River and Baiyangdian Lake

Field hydrochemical parameters for upstream, midstream, downstream, and Baiyangdian Lake are summarized in . From 2008 to 2016, pH values ranged from 7.4 to 8.2 in the Fuhe River and 8.1 to 8.6 in Baiyangdian Lake, indicating weakly alkaline conditions. Interannual variations in average EC and TDS followed the order: June 2009 > June 2011 > July 2014 > September 2008 > June 2016, with consistent patterns observed along the river course. TDS and EC exhibited clear seasonal trends: pre-rainy season (June 2009 and 2011) > rainy season (July 2014) > post-rainy season (September 2008), demonstrating precipitation' s dominant influence on water quality. Notably, despite similar pre-rainy season timing, TDS and EC in June 2016 were lower than in 2009 and 2011, likely reflecting changes in sewage discharge volumes, concentrations, or altered water use patterns in Baoding following the South-to-North Water Diversion Project. Baiyangdian Lake showed smaller EC and TDS fluctuations due to its larger water volume and capacity, maintaining relatively stable water quality compared to the Fuhe River, which exhibited substantial interannual and intra-annual variations driven by precipitation dilution, discharge control, and self-purification processes.

Piper diagrams reveal diverse water chemistry types in the Fuhe River ([Figure 2: see original paper]). In 2008, types included $Na \cdot Ca\text{-}Cl \cdot SO_4 \cdot HCO_3$, $Na \cdot Ca\text{-}SO_4$, and $Na \cdot Ca\text{-}SO_4 \cdot HCO_3$, with elevated SO_4^{2-} from non-ferrous metal smelting wastewater. In 2009, the river showed $Na \cdot Ca\text{-}HCO_3 \cdot SO_4$ type, also indicating sulfide- or sulfate-rich industrial wastewater input. The 2011 chemistry was predominantly $Na \cdot Ca\text{-}Cl \cdot HCO_3$ type, shifting to $Na \cdot Ca\text{-}HCO_3 \cdot Cl$ in 2014 and $Na \cdot Ca \cdot Mg\text{-}HCO_3$ in 2016. Baiyangdian Lake exhibited $Na \cdot Ca\text{-}HCO_3 \cdot Cl \cdot SO_4$ type in 2008, $Na \cdot Mg\text{-}HCO_3 \cdot Cl \cdot SO_4$ in 2009, $Na \cdot Mg\text{-}HCO_3 \cdot Cl$ in 2014, and $Na \cdot Ca \cdot Mg\text{-}HCO_3 \cdot Cl$ in 2016. The consistent interannual evolution of water chemistry types between the Fuhe River and Baiyangdian Lake demonstrates the river' s direct influence on lake water quality, while differences reflect transformations occurring along the flow path and impacts from local residents and agricultural activities. Notably, the proportions of Cl^- and SO_4^{2-} gradually decreased from 2008 through 2014, indicating improved control over industrial and domestic discharges.

2.2 Hydrogen and Oxygen Isotope Characteristics

Hydrogen and oxygen isotope values for both water bodies plot below the Global Meteoric Water Line and Local Meteoric Water Line ([Figure 3: see original paper]), with varying distances from the local line across years and seasons. This indicates that Fuhe River water originates primarily from precipitation, but with different seasonal sources and evaporation intensities. Baiyangdian Lake shows more enriched δ^2H and $\delta^{18}O$ values than the Fuhe River, reflecting stronger evaporative enrichment. Seasonal patterns among pre-rainy (June 2011), rainy (July 2014), and post-rainy (September 2008) seasons reveal progressive isotopic enrichment along the river, consistent with the seasonal effect where early rainfall contains more enriched isotopes. Given the river' s 6-day renewal time, the isotopic signatures represent mixing of sewage discharge and precipitation recharge. Stable domestic water sources in June 2011, July 2014, and September 2008 resulted in isotopic distributions reflecting both seasonal precipitation patterns and varying evaporation. The absence of a clear seasonal pattern in June 2016 likely stems from altered water sources in Baoding following the South-to-North Water Diversion Project' s commissioning in December 2014 [18]. Except for 2008, longitudinal trends in 2011, 2014, and 2016 show progressive downstream isotopic enrichment, indicating enhanced evaporation along the flow path.

2.3 Spatiotemporal Variations and Source Identification of Nitrate

Mean nitrate concentrations in the Fuhe River followed the order: June 2009 (85.0 $\mathrm{mg \cdot L^{-1}}$) > June 2011 (13.7 $\mathrm{mg \cdot L^{-1}}$) > July 2014 (6.3 $\mathrm{mg \cdot L^{-1}}$) ([Figure 4: see original paper]). This interannual variation confirms that besides precipitation dilution, industrial wastewater discharge in 2009 caused extreme nitrate concentrations exceeding the WHO drinking water standard of 50 $\mathrm{mg \cdot L^{-1}}$ at all sites. Baiyangdian Lake showed mean NO_3^- concentrations of 7.02 $\mathrm{mg \cdot L^{-1}}$ (2008), 5.67 $\mathrm{mg \cdot L^{-1}}$ (2009), 0.79 $\mathrm{mg \cdot L^{-1}}$ (2014), and 2.61 $\mathrm{mg \cdot L^{-1}}$ (2016)—substantially lower than Fuhe River values. The upstream-midstream-downstream pattern of increasing then decreasing NO_3^- concentrations indicates that besides self-purification during flow, dilution by external recharge, biological uptake, and denitrification within the lake also influence water quality.

Potential nitrate sources include atmospheric deposition, industrial wastewater, animal manure, domestic sewage, and soil microbial nitrogen. According to Xue et al. [20], domestic sewage $\delta^{15}N$ ranges from 4‰ to 19‰, while industrial wastewater ranges from -4‰ to 15‰. Chloride, derived primarily from domestic sewage and unaffected by physical, chemical, or microbial processes, serves as a key indicator of wastewater influence. In 2008, $\delta^{15}N$ values >10‰ indicated domestic sewage as the primary nitrate source ([Figure 5a: see original paper]). The 2009 range of -3.7‰ to 4.0‰ confirmed industrial wastewater dominance, explaining the anomalous nitrate exceedances that year and highlighting industrial discharge' s severe impact. Site F1-2009, located at Yindingzhuang upstream, was a domestic sewage channel with high Cl^- and NO_3^- concentrations ([Figure 5b: see original paper]). In 2014, $\delta^{15}N$ values of 2.07‰-18.49‰ with high Cl^- concentrations indicated domestic sewage sources, with site F2-2014 at Xianrenqiao Village representing background conditions unaffected by wastewater discharge.

Baiyangdian Lake $\delta^{15}N$ values of 5.8‰-11.7‰ (2008) and 3.31‰-12.53‰

(2014) reflected diverse nitrate sources, including Fuhe River domestic sewage, local residents, and agricultural fertilizer application. The 2009 range of -3.8‰ to 0.7‰ demonstrated direct industrial wastewater impacts from the Fuhe River.

2.4 Factors Controlling Nitrate Migration and Transformation

Nitrate concentrations are controlled by new source injection, dilution, and biochemical reactions. Dissolved oxygen (DO) is a critical parameter characterizing redox conditions. In June 2016, measured NO_3^- concentrations exceeded calculated values while showing downstream decreasing trends, with DO ranging from 0.3 to 1.1 $mg \cdot L^{-1}$, indicating anoxic conditions. This suggests denitrification dominated nitrate reduction along the river course ([Figure 6a: see original paper]). In June 2009, all measured NO_3^- values were lower than calculated values, with biochemical reactions controlling concentrations. The downstream sequence F1 (NO_3^-, 130.8 $mg \cdot L^{-1}$) → F2 (NO_3^-, 72.2 $mg \cdot L^{-1}$) → F3 (NO_3^-, 71.4 $mg \cdot L^{-1}$) showed decreasing nitrate with enriched $\delta^{15}N$ values (F1: -3.7‰ → F2: -2.0‰ → F3: 4.0‰), reflecting denitrification under anoxic conditions induced by combined domestic and industrial sewage inputs.

In June 2011, despite isotopic enrichment indicating enhanced evaporation, measured NO_3^- exceeded calculated values, suggesting external inputs controlled nitrate variation. July 2014 showed increased nitrate from F3 (5.4 $mg \cdot L^{-1}$) to F4 (14.8 $mg \cdot L^{-1}$), with measured values exceeding calculated values, indicating additional nitrate inputs along this reach. From F6 (8.8 $mg \cdot L^{-1}$) to F7 (4.6 $mg \cdot L^{-1}$) in 2014, decreasing nitrate with measured values below calculated values indicated biochemical reaction control, likely denitrification. This interpretation is supported by DO < 2 $mg \cdot L^{-1}$ at F6 and F7, and increased HCO_3^- from 459.0 to 487.5 $mg \cdot L^{-1}$, consistent with the denitrification reaction (Equation 2):

$$5/4CH_2O + NO_3^- + 1/4H^+ \rightarrow 1/2N_2 + 5/4HCO_3^- + 1/2H_2O \qquad (2)$$

Conclusion

Analysis of hydrochemical characteristics and water types from 2008-2016 revealed that precipitation controls total ion concentrations, with TDS and EC in the Fuhe River following the pattern: pre-rainy season > rainy season > post-rainy season. Baiyangdian Lake maintained more stable water quality due to its larger volume. The gradual decrease in Cl^- and SO_4^{2-} proportions from 2008-2014 indicated successful control of industrial and domestic discharges. The Fuhe River' s water chemistry evolution directly influenced Baiyangdian Lake, while differences reflected local impacts and transformation processes.

δ^{15}N analysis confirmed that nitrate pollution in the Fuhe River originated primarily from village domestic sewage, with minimal agricultural non-point source contribution, except for June 2009 when industrial wastewater caused severe exceedances. Baiyangdian Lake δ^{15}N values mirrored Fuhe River patterns, confirming the river as the main nitrate source, while wider δ^{15}N ranges reflected additional contributions from local residents and agricultural fertilizers. Precipitation dilution and external inputs were the primary controls on nitrate variation along the river-lake continuum, while denitrification dominated nitrate reduction when DO < 2 mg $\cdot$ L^{-1} in June 2009, July 2014, and June 2016. Effective control of domestic and industrial discharges from the Fuhe River is therefore essential for managing nitrate pollution in Baiyangdian Lake.

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Note: Figure translations are in progress. See original paper for figures.