ISSN: 2710-0723

Spatial Heterogeneity and Vertical Distribution of Nitrogen Forms in Sediments of Qilu Lake

Wei Li[1,2], Tianxiang Li[2], Mingyan Liu[2,4], Yaping Liu[2], Lixin Jiao[2], Miao Zhang[3], Bo Yao[3], Jingyi Yan[3], Jianxun Wu[3], Xin Wen[3], and Dengling Jiang[1,*]

[1] North China University of Science and Technology, Tangshan 063210, China

[2] State Key Laboratory of Environmental Criteria and Risk Management, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

[3] Kunming Research Academy of Ecological and Environmental Sciences, Kunming 650032, China

[4] Xinjiang Tianhe Environmental Technology Consulting Co., Ltd., Urumqi 830092, China

*Corresponding Author

Abstract

To reveal the spatial heterogeneity and vertical distribution characteristics of nitrogen forms in the sediments of Qilu Lake, overlying water and surface sediment samples were collected from 15 sampling sites in June 2023, and sediment core samples were additionally collected from seven representative sites. The occurrence characteristics, release contributions, and interrelationships of various nitrogen fractions in overlying water, surface sediments, and sediment cores were systematically analyzed. The results showed that inorganic nitrogen dominated the nitrogen pool in the overlying water of Qilu Lake, with relatively low NH_4^+-N concentrations and comparatively higher NO_3^--N concentrations. Site Q9 exhibited the most pronounced nitrogen enrichment. DIN was the predominant component of DTN. In surface sediments, TN, PMON, SN, and various forms of EN and PMN all exhibited marked spatial heterogeneity, and the high-value zones of different nitrogen pools did not completely overlap, indicating that various nitrogen fractions possess distinct distribution patterns within the lake basin. The release contributions of nitrogen forms in sediment cores differed significantly among lake zones, with the western zone showing the most pronounced inter-site variability. Vertical profiles of sediment cores revealed that NH_4^+-N concentrations were relatively high in shallow layers at most sites, whereas site Q9 exhibited a distinct increasing trend with depth. NO_3^--N concentrations fluctuated considerably among sites. PMN generally decreased from shallow to deep layers. TN remained relatively stable with depth at most sites, with only site Q15 showing notable vertical variation. Correlation analysis indicated that, among the labile nitrogen fractions in surface sediments, NH_4^+-N in EN exhibited the strongest correlations with overlying water NH_4^+-N, DIN, and DTN, while other labile nitrogen fractions showed relatively weaker correlations with inorganic nitrogen in overlying water. These findings demonstrate that nitrogen fractions in the sediments of Qilu Lake exhibit significant heterogeneity in both spatial and vertical dimensions, and that there exists a strong coupling relationship between the surface labile nitrogen pool and inorganic nitrogen in the overlying water, which can provide a scientific basis for the identification and management of internal nitrogen pollution in the lake.

Keywords

Qilu Lake; Sediment; Nitrogen Forms; Vertical Distribution.

ISSN: 2710-0723

1. Introduction

The quality and sustainable management of water resources represent a major global challenge, particularly for freshwater lakes. Freshwater lakes not only serve as critical habitats for biodiversity but also constitute vital resources for human socio-economic development[1]. In China, eutrophication has been a prominent issue for freshwater lakes since the economic reforms of the 1980s, with harmful cyanobacterial blooms occurring in drinking water sources and recreational areas[2]. Although a series of laws and regulations on water pollution prevention have improved overall water quality, lake eutrophication has not been effectively reversed. Qilu Lake, a typical freshwater lake located in Tonghai County, Yuxi City, on the Yunnan–Guizhou Plateau, is subject to a heavy burden as 90% of the local population resides within its basin. The lake has been severely polluted since the mid-to-late 1980s [3]and has long remained in a state of water quality worse than Grade V, with nitrogen pollution being particularly prominent. Nitrogen, as a key nutrient, can cause eutrophication when input in excess, leading to excessive algal growth and blooms, which not only disrupt the ecological balance of the water body but also seriously impair water quality and aquatic organism health. By analyzing nitrogen forms in sediments, the major pollution sources-such as domestic sewage [4]and agricultural wastewater [5]-can be identified, thereby providing a basis for formulating effective pollution control and remediation strategies.

Due to the severe eutrophication of Qilu Lake, a large number of researchers have conducted studies on this lake. Some scholars have analyzed eutrophication through water environment investigations. Qin et al. [6]studied the water quality and eutrophication status of Qilu Lake and found that total nitrogen in the water column was the most significant environmental factor contributing to the eutrophic state, while phosphorus was the limiting factor for algal growth and reproduction. Zhou [7]carried out monthly monitoring of various water quality parameters in Qilu Lake from November 2016 to October 2017, and further explored the ecological conditions and monthly variations in water quality. The results indicated that the lake was in a moderate to severe eutrophic state during the monitoring period, with nitrogen and phosphorus concentrations remaining at high levels throughout the year; agricultural discharge had a greater impact on total nitrogen than on total phosphorus. In recent years, researchers have begun to study Qilu Lake through aquatic organisms. Du et al. [8]conducted four surveys of phytoplankton and eutrophication-related water quality parameters in 2021, and assessed the fish productivity of the lake. Their calculations revealed a mean Trophic Level Index (TLI) of 76.3, indicating a severe eutrophic state. Reducing external inputs of nutrients such as nitrogen and phosphorus into inflowing rivers remains one of the primary measures for managing eutrophication in typical lakes. However, simply reducing or blocking external loads cannot completely reverse lake eutrophication, because internal release and recycling of sediment nitrogen can maintain a prolonged eutrophic state even after substantial reduction of external loads[9,10], thereby hindering lake restoration.

To date, research on nitrogen and phosphorus in the sediments of Qilu Lake has been relatively limited. Yin et al. [11]analyzed the contents of organic matter (OM), total nitrogen (TN), and total phosphorus (TP) as well as grain-size parameters in surface sediments of Qilu Lake, and evaluated the pollution levels. The nutrient pollution assessment indicated that OM pollution was relatively low, whereas TN and TP pollution were more severe. Chen et al. [12]investigated the variations in dissolved organic nitrogen (DON) and amino acids (AA) in the sediments of Qilu Lake, and explored the molecular weight fractionation characteristics of DON. They found that the concentrations of DON and AA were relatively high in the severely polluted lake, mainly derived from domestic sewage input from villages built along the lake, and were significantly influenced by human activities. To date, studies on eutrophication in Qilu Lake have largely focused on the above aspects, with most researchers taking the lake water environment as the

ISSN: 2710-0723

primary subject (neglecting internal source pollution), while sediment research has remained mainly at the level of surface TN, TP, and OM (lacking studies on the vertical distribution of nitrogen forms in sediments). Therefore, this paper investigates the spatial and vertical distribution characteristics of nitrogen forms in the sediments of Qilu Lake and their driving factors, based on the occurrence characteristics of nitrogen forms in overlying water and surface sediments from 15 sampling sites, as well as vertical analyses of nitrogen forms in sediment cores from 7 sites. In addition, the contributions of nitrogen release from different lake zones are analyzed, with the aim of providing a scientific basis for the management of lake eutrophication.

2. Materials and Methods

2.1. Study Area

Qilu Lake (24°12′–24°0′ N, 102°45′–120°48′ E) is located in Tonghai County, Yuxi City, Yunnan Province, and belongs to the Xijiang River system of the Pearl River Basin. The lake covers an area of 36.86 km^2, with a maximum depth of 6.8 m and an average depth of 4.0 m[13]. It is one of the nine major plateau lakes in Yunnan. In June 2023, sampling was conducted at 15 sites for overlying water and surface sediments, and at 7 of these sites for sediment cores. The lake was divided into different zones based on latitude, longitude, and water depth; the specific sampling sites and lake zones are shown in Fig. 1. Overlying water was collected using a plexiglass water sampler, with minimal disturbance to the sediments, and the water samples were stored in pre-cleaned polyethylene bottles. Surface sediments were collected using a Peterson grab sampler to obtain the top 10 cm of sediment, which was then placed in insulated containers. Sediment cores were collected using a gravity corer (9 cm inner diameter). Prior to analysis, the sediment cores were sliced into intervals of 0–1, 1–2, 2–4, 4–6, 6–8, 8–10, 10–12, 12–16, 16–20, 20–24, 24–28, and 28–32 cm. The sliced samples and surface sediment samples were freeze-dried at –80 °C for 7 days in sterile plastic bags, ground, and passed through a 100-mesh sieve before being used for nitrogen analysis.

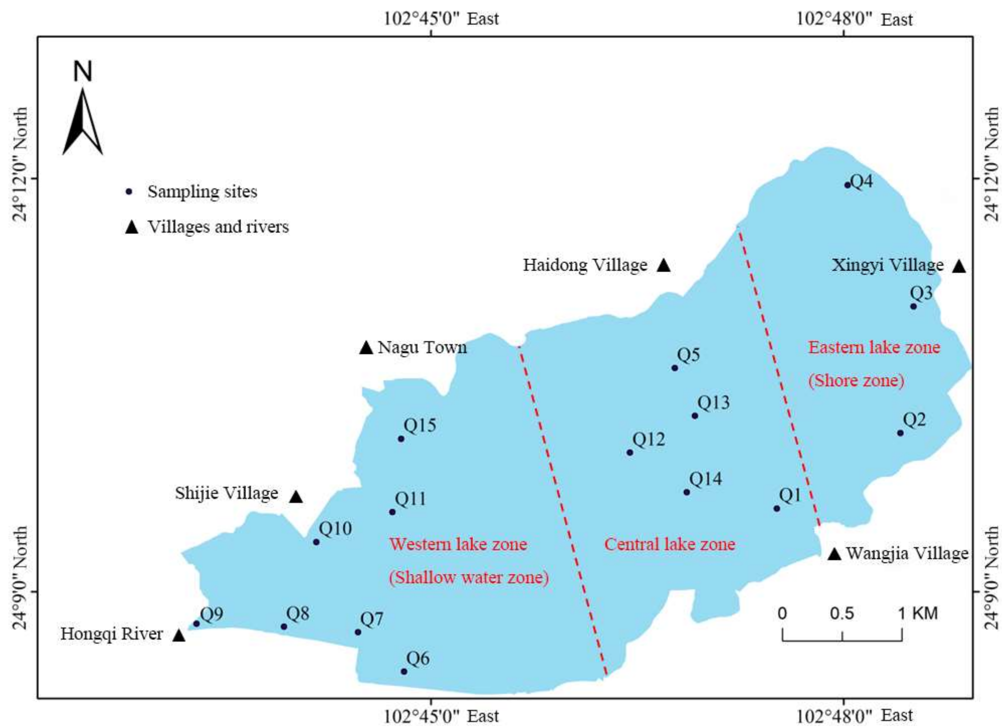


Fig. 1 Locations of sampling sites in Qilu Lake.

ISSN: 2710-0723

2.2. Experimental Methods

After freeze-drying, the sediment sub-samples were ground in an agate mortar and passed through a 100-mesh sieve. Total nitrogen (TN) in the sediments was determined using the alkaline potassium persulfate digestion method. Exchangeable nitrogen (EN, exchangeable nitrogen) was extracted as follows: 5 g of the sieved sediment sample was mixed with 60 mL of 0.5 mol·L^{-1} K_2SO_4 solution in a 100 mL polyethylene centrifuge tube. The mixture was shaken at 200 r·min^{-1} for 30 min and then centrifuged at 6000 × g for 10 min. The contents of NO_3^--N, NO_2^--N, and NH_4^+-N in the extract were determined by ultraviolet spectrophotometry, diazo-azo spectrophotometry, and Nessler's reagent spectrophotometry, respectively. Potential mineralizable nitrogen (PMN, potential mineralizable nitrogen) was determined by incubating 5 g of sediment samples at 40 °C for 7 days, followed by extraction with K_2SO_4 solution, according to the procedures described in the *Standard Methods for the Examination of Water and Wastewater* (4th edition)[14]. Potential mineralizable organic nitrogen (PMON) is the organic nitrogen fraction that can be microbially mineralized and transformed in sediments, and was calculated by the waterlogged incubation difference method, i.e., PMON was obtained as the difference between the total inorganic nitrogen after incubation and the exchangeable nitrogen. Stable nitrogen (SN) was calculated by difference: SN = TN − EN − PMON. The classification and calculation of PMON and SN followed Ding et al.[23]. All experiments were performed in triplicate, and the results were expressed as mean values with an error margin controlled within 5%.

2.3. Calculation of Nitrogen Release Contribution

The release contribution equation [15]can be expressed as:

$$Release\ contribution = \frac{|C_{surface} - C_{other}|}{\Sigma_{i=1}^{n}|C_{surface} - C_{other}|} \times 100\% \quad (1)$$

Where $C_{surface}$ represents the content of nitrogen forms in the upper sediment layer, and C_{other} represents the concentration of nitrogen forms in the other sediment layers. If the difference between $C_{surface}$ and C_{other} is positive, the substances are mainly released; conversely, they are mainly retained.

2.4. Statistical Analysis

All experimental data were compiled and processed using Microsoft Excel 2019. Correlations among different variables were analyzed using Pearson's correlation coefficient. Statistical analyses were performed with SPSS 22.0. Figures were plotted and integrated using Origin 9.1, ArcGIS 10.8, and R 4.5.0.

3. Results and Analysis

3.1. Basic Water Quality Indicators of Overlying Water in Qilu Lake

Table 1 presents the concentrations of basic water quality indicators in the overlying water at each sampling site in Qilu Lake. Overall, inorganic nitrogen dominated the nitrogen pool in the overlying water, and different nitrogen forms exhibited distinct spatial variations within the lake. The COD_{Mn} concentration in the overlying water ranged from 12.0335 to 24.6531 mg·L^{-1}, showing relatively small variations; while COD_{Cr} varied more widely, ranging from 28.2818 to 150.7358 mg·L^{-1}, with the highest value observed at Site Q5. NH_4^+-N concentrations ranged from 0.1052 to 5.3634 mg·L^{-1}, with most sites below 1 mg·L^{-1}. NO_3^--N concentrations were generally higher than NH_4^+-N, ranging from 1.3876 to 9.6183 mg·L^{-1}, and concentrated between

ISSN: 2710-0723

1.89 and 3.3295 mg·L^{-1} at most sites. The concentrations of DIN, DTN, and TN ranged from 1.5899 to 14.9817 mg·L^{-1}, 1.5915 to 15.6808 mg·L^{-1}, and 1.9996 to 16.1823 mg·L^{-1}, respectively, and their spatial patterns were generally consistent with that of NO_3^--N.

Spatially, Site Q1 generally showed low nitrogen concentrations, whereas Site Q9 exhibited the highest values of NH_4^+-N, NO_3^--N, DIN, DTN, and TN across the entire lake, indicating pronounced local enrichment. Sites Q6–Q9 were identified as high-value areas for NO_3^--N, with Site Q9 being the most prominent. Except for Site Q5, the high-value areas for DIN generally coincided with those for NO_3^--N, suggesting that the inorganic nitrogen composition in the overlying water was mainly controlled by NO_3^--N variations. The high-value sites for DTN and TN were similarly distributed, indicating that dissolved components accounted for a substantial proportion of total nitrogen during the sampling period.

In terms of compositional ratios, DIN predominated in DTN, with DIN/DTN exceeding 85% at most sites. DTN also constituted a high proportion of TN, with DTN/TN above 80% at most sites. However, DIN/DTN at Sites Q5 and Q15 were relatively lower, at 58.11% and 65.73%, respectively, while the lowest DTN/TN (55.57%) was found at Site Q14. Phosphorus concentrations in the overlying water were generally lower than nitrogen, with TDP and TP ranging from 0.0161 to 0.5223 mg·L^{-1} and 0.0779 to 0.7272 mg·L^{-1}, respectively. Nevertheless, Site Q5 still exhibited relatively high phosphorus levels. Overall, based on all indicators, Site Q5 showed a relatively higher pollution load in the overlying water, while Site Q9 exhibited more prominent nitrogen enrichment.

Table 1. Concentrations of water quality indicators in the overlying water of Lake Qiluhu at various sites

mg·L^{-1}

sampling site	COD_{Mn}	COD_{Cr}	NH_4^+-N	NO_3^--N	DIN	DTN	TN	DTP	TP
Q1	12.0335	44.2154	0.1064	1.8900	1.9964	2.2250	2.2872	0.0432	0.0779
Q2	16.9782	53.2253	1.0436	2.1233	3.1669	3.4923	3.9441	0.0686	0.1364
Q3	15.2219	47.3550	0.5808	1.8190	2.3998	3.1007	3.1017	0.0506	0.0642
Q4	11.5988	44.7634	1.0124	1.7064	2.7188	3.1000	3.4901	0.1057	0.1104
Q5	24.6531	150.7358	4.4212	3.3295	7.7507	13.3373	14.5489	0.5520	0.7272
Q6	15.5630	32.5152	0.2434	7.0134	7.2568	7.8356	8.5346	0.1923	0.2132
Q7	15.1045	52.2016	0.4131	6.3766	6.7897	7.2338	7.8253	0.0219	0.1041
Q8	15.0179	39.5159	2.0675	5.2178	7.2853	7.6895	8.2587	0.5223	0.5024
Q9	15.8270	28.2818	5.3634	9.6183	14.9817	15.6808	16.1823	0.0592	0.1165
Q10	15.0548	38.1178	0.1052	1.8110	1.9162	2.0062	2.2529	0.0282	0.0790
Q11	12.2011	38.6859	0.1780	1.6482	1.8262	1.8408	2.1787	0.0182	0.0760
Q12	13.2730	40.9469	0.2023	1.3876	1.5899	1.5915	1.9996	0.0438	0.0588
Q13	20.8081	97.7508	0.3667	2.5103	2.8770	3.3509	4.5913	0.1181	0.3055
Q14	20.5931	73.9511	0.2084	1.9356	2.1440	2.1554	3.8785	0.0351	0.2693
Q15	12.4476	45.515	0.0845	1.5935	1.6780	2.5527	3.1778	0.0161	0.1179

3.2. Spatial Distribution Characteristics of Nitrogen Forms in Surface Sediments of Qilu Lake

The spatial distributions of different nitrogen fractions in the surface sediments of Qilu Lake are shown in Fig. 2. Overall, TN, PMON, SN, and various forms of EN and PMN all exhibited certain degrees of spatial heterogeneity, but the high-value and low-value areas of different fractions did not completely overlap, indicating that the occurrence patterns of different nitrogen pools in the surface sediments were distinctly different.

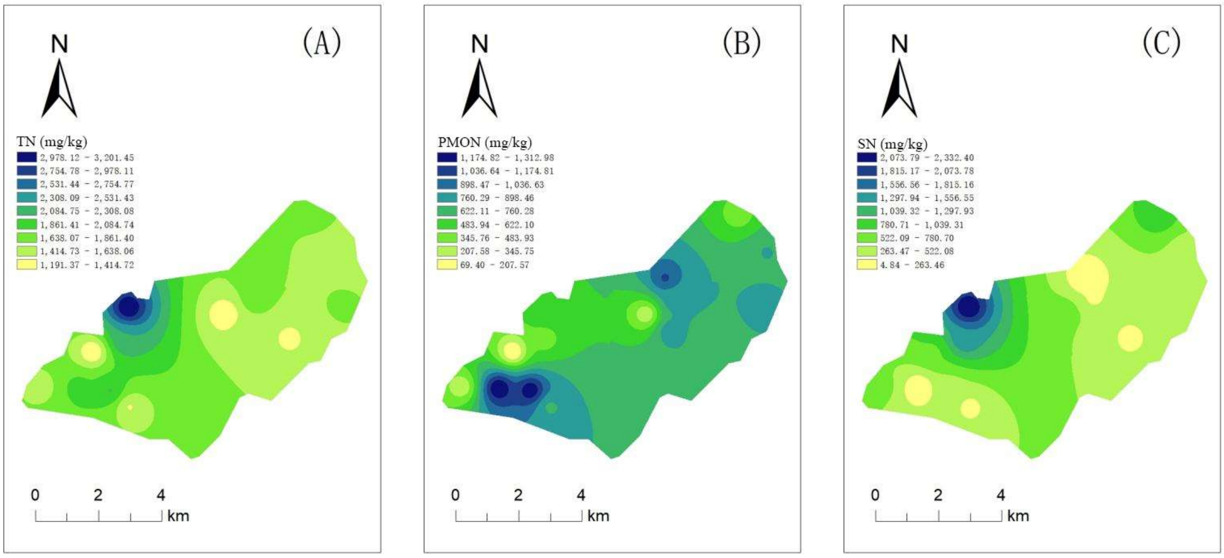
ISSN: 2710-0723

Total nitrogen in the surface sediments was generally at a relatively high level, with a spatial distribution characterized by localized high values and relatively moderate values in the remaining areas. The high-value areas were mainly found in the northwestern part of the lake, while the lake center and parts of the eastern area showed relatively lower values, suggesting that TN in the lake exhibited a spatial pattern of both local enrichment and overall moderate uniformity. Compared with TN, the spatial patterns of PMON and SN showed certain differences. PMON exhibited relatively high values in the western side and parts of the central lake area, indicating that these areas had higher reserves of potential mineralizable organic nitrogen. SN formed a pronounced high-value center in the northwestern part of the lake, while the remaining areas were generally low, suggesting that the distribution of stable nitrogen varied more concentratedly across different lake zones.

Among the exchangeable inorganic nitrogen fractions, NH_4^+-N in EN and NO_3^--N in EN exhibited different spatial distribution characteristics. High values of NH_4^+-N in EN were mainly located in the western side of the lake, while the rest of the lake showed relatively low concentrations with a relatively flat distribution. The spatial heterogeneity of NO_3^--N in EN was more pronounced, with high values mainly concentrated in the northeastern and north-central parts of the lake, and relatively lower values in the southwestern part and parts of the central lake area, indicating that exchangeable NO_3^--N exhibited stronger site-specific variability within the lake.

Among the potentially mineralizable inorganic nitrogen fractions, both NH_4^+-N in PMN and NO_3^--N in PMN showed considerable spatial variation, but their distribution patterns were not consistent. High values of NH_4^+-N in PMN were mainly concentrated in the western side of the lake, with a secondary high-value center in the north-central area. NO_3^--N in PMN exhibited relatively high values in the southwestern and north-central parts of the lake, while relatively lower values were found in the eastern part and parts of the central lake area. Overall, the high-value areas of different inorganic nitrogen forms in PMN showed a certain degree of interleaving, indicating that the composition of potential mineralization products in surface sediments differed markedly among lake zones.

In summary, the spatial distributions of the total nitrogen pool, organic nitrogen pool, and labile inorganic nitrogen pool in the surface sediments of Qilu Lake were not synchronized. TN and SN exhibited more pronounced enrichment in localized areas, while different inorganic nitrogen forms in EN and PMN showed greater spatial variability, suggesting that the occurrence, transformation, and potential release risks of nitrogen in surface sediments across the lake exhibited significant spatial heterogeneity.



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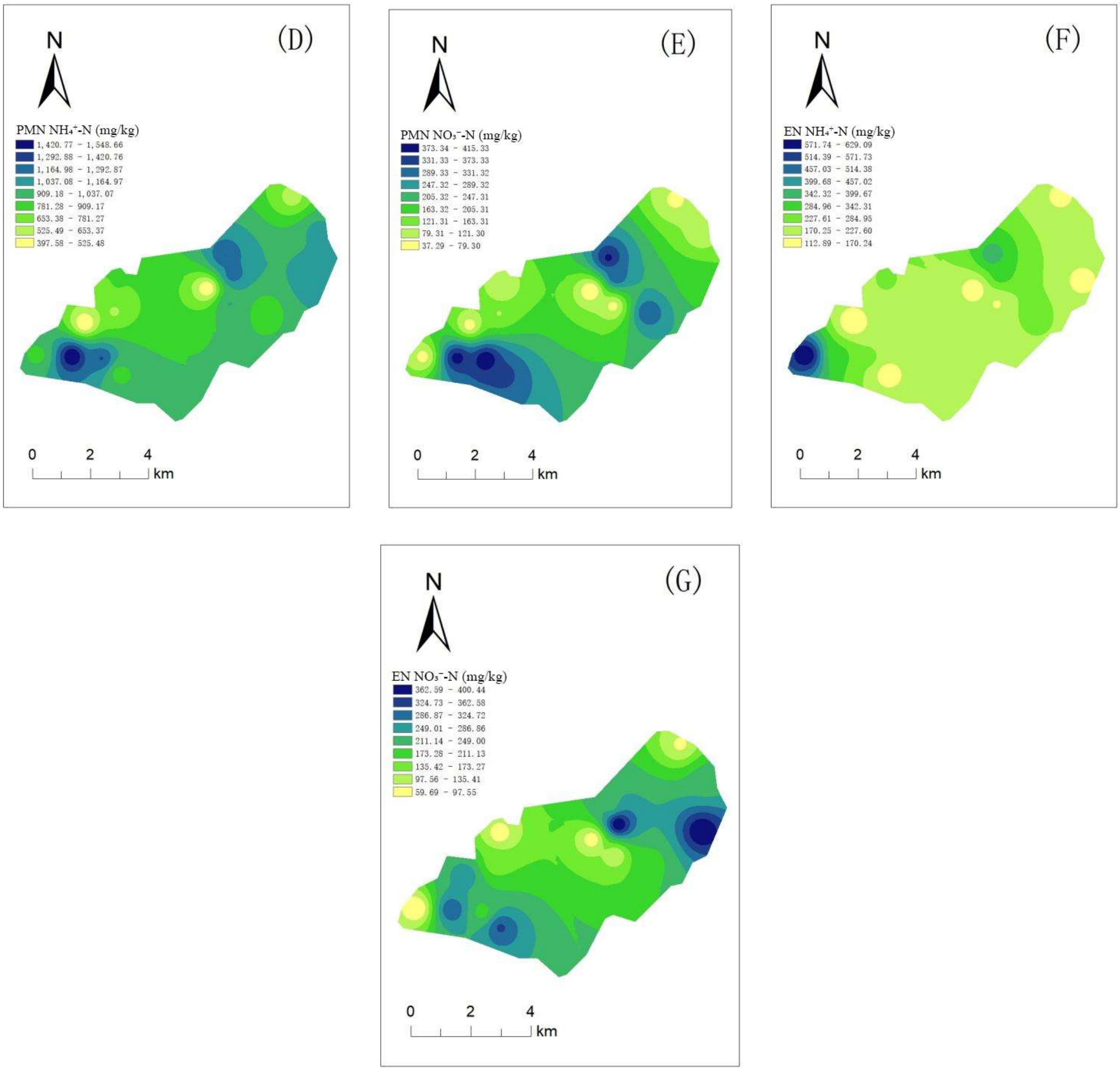

Fig. 2 Spatial distribution of nitrogen patterns in surface sediments of Lake Qiluhu

3.3. Release Contribution of Sedimentary Nitrogen in Different Lake Zones of Qilu Lake

In this section, sediment cores of 0–32 cm depth were selected from seven sites in the western (Q9, Q10, Q15), central (Q12, Q14), and eastern (Q3, Q4) zones of Qilu Lake, with cores from two sites not reaching 32 cm (Q10: 0–20 cm; Q12: 0–24 cm) but still adequately reflecting the release contributions and vertical distribution characteristics, and the cores were divided into shallow (0–10 cm), middle (11–20 cm), and deep (21–32 cm) layers according to depth intervals. The release contributions of nitrogen forms in the sediments of Qilu Lake across different lake zones and depths are shown in Fig. 3; the release contributions of nitrogen forms at different depths remained relatively stable, however, notable differences were observed among lake zones. In the western zone, the release contributions varied among sites: at Q9, NH_4^+-N accounted for the highest proportion (59.3%–71.6%) while NO_3^--N had the lowest (2.4%–2.8%); at Q15, the NH_4^+-N contribution was slightly lower (17%–18.9%) and PMN dominated with the highest contribution (72.5%–74.2%); at Q10, the NO_3^--N contribution was the highest among the western sites (37.9%–46.5%) and PMN showed a similar range (37.6%–44%). In the central zone, PMN dominated the release contribution at both sites, accounting for

ISSN: 2710-0723

over 80% with similar values, however, exchangeable nitrogen fractions exhibited considerable differences: at Q14, NH_4^+-N contributed 3.1%–4.9% and NO_3^--N contributed 8.6%–11.8%, whereas at Q12, the opposite pattern was observed, with NH_4^+-N contributing 10%–11.1% and NO_3^--N contributing only 2.3%–2.5%. In the eastern zone, the release contributions were relatively stable, with PMN consistently accounting for the highest proportion (over 65%), and at Q3, PMN was even higher (83.2%–88.9%) while NH_4^+-N showed the lowest contribution, with a more pronounced effect at Q3 (3.3%–5%).

Fig. 3 Contribution of nitrogen form release from Lake Qiluhu sediments

3.4. Vertical Distribution Characteristics of Nitrogen Forms in Sediments of Qilu Lake

The vertical distributions of NH_4^+-N, NO_3^--N, PMN, and TN in the seven sediment cores from Qilu Lake are shown in Fig. 4. Overall, the vertical variation characteristics of these nitrogen fractions differed markedly among the cores, with NH_4^+-N and PMN exhibiting relatively clear inter-layer differentiation in most cores, while NO_3^--N and TN showed greater differences in both variation amplitude and stability across different sites.

As shown in Fig. 4(A), with the exception of Q9, the NH_4^+-N concentrations in the remaining six cores showed relatively small variations with depth, characterized mainly by slight fluctuations vertically, with most cores exhibiting relatively higher concentrations in the shallow layers and lower concentrations in the middle and deep layers. The fluctuation amplitude at Q15 was relatively larger, but the overall pattern still featured higher concentrations in the surface layer. Unlike the other sites, the NH_4^+-N concentration in the Q9 core increased markedly with depth, with deep-layer concentrations significantly higher than those in the surface layer, making it the most prominent core among the seven in terms of vertical variation.

As shown in Fig. 4(B), the vertical distribution of NO_3^--N was more unstable than that of NH_4^+-N, and the differences among the cores were also more pronounced. The NO_3^--N concentrations

ISSN: 2710-0723

at Q10, Q14, and Q15 fluctuated considerably with depth, with notable high-value variations in some layers; while the vertical changes at Q3, Q4, Q9, and Q12 were relatively gentle, with Q9 generally showing low concentrations and small inter-layer differences. Overall, NO_3^--N was relatively high in the surface layers of some cores, but no consistent unidirectional trend was observed in the middle and deep layers.

As shown in Fig. 4(C), PMN concentrations in the seven cores were significantly higher than EN fractions, and the surface layer contents were higher than those in the deep layers in most cores. The vertical variation patterns of PMN at Q3, Q4, Q9, and Q12 were relatively similar, generally showing a gradual decrease with depth; Q14 and Q15 showed larger variation amplitudes, with Q15 exhibiting some fluctuations in local surface layers; the vertical variation of PMN at Q10 was relatively moderate, characterized by an overall increase followed by a decrease. Overall, the enrichment characteristics of PMN in shallow sediments were relatively obvious.

As shown in Fig. 4(D), the vertical variation of TN across the different cores was generally weaker than that of PMN. At Q3, Q4, Q9, Q10, Q12, and Q14, TN concentrations remained relatively stable with depth, with no consistent trend of significant increase or decrease. In contrast, the vertical variation of TN at Q15 was the most pronounced, with overall higher concentrations in the shallow and middle layers and lower concentrations in the deep layer. This indicates that there were some differences in TN accumulation characteristics among cores from different lake zones, but the vertical distribution of TN at most sites remained relatively stable.

Overall, as shown in Figs. 4(A)–(D), the vertical distributions of different nitrogen fractions in the sediment cores from Qilu Lake were not synchronous. NH_4^+-N and PMN exhibited relatively surface enrichment in most cores, while NO_3^--N showed greater inter-layer fluctuations, and TN was generally stable at most sites, with only individual cores showing notable vertical differences.

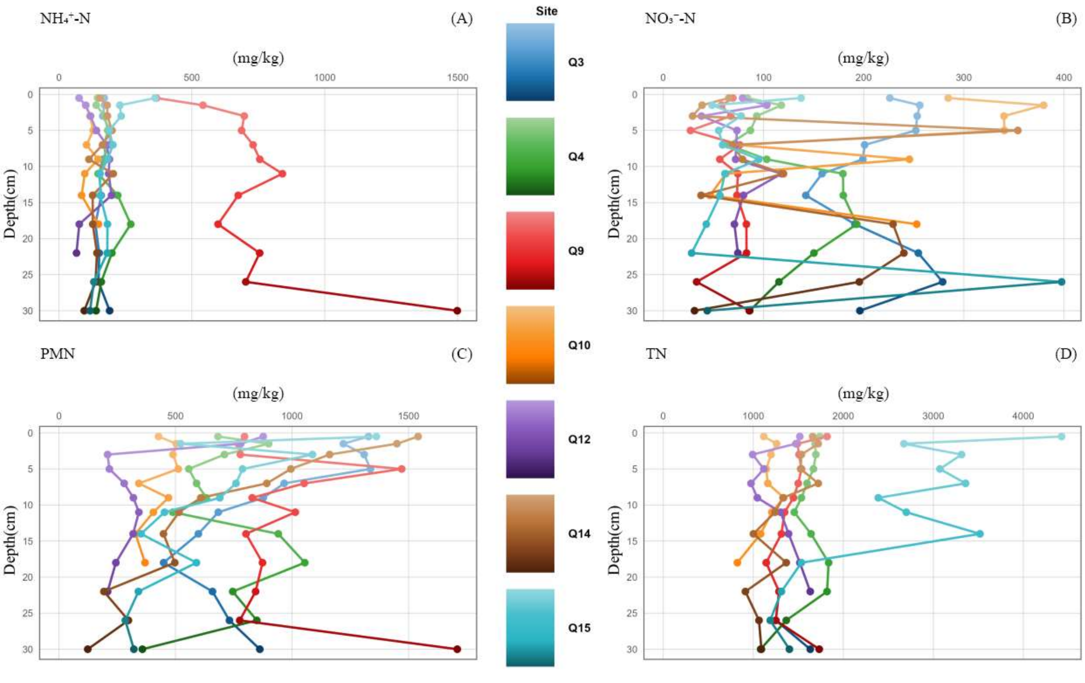

Fig. 4 Vertical profiles of NH_4^+-N, NO_3^--N, PMN, and TN in seven sediment cores from Lake Qiluhu

ISSN: 2710-0723

4. Discussion

4.1. Relationship between Labile Nitrogen Fractions in Surface Sediments and Inorganic Nitrogen in Overlying Water in Qilu Lake

Fig. 5 shows the Pearson correlation between labile nitrogen fractions in surface sediments and inorganic nitrogen in overlying water in Qilu Lake. Overall, the correlation strengths between different labile nitrogen fractions and various inorganic nitrogen indicators in overlying water differed markedly, among which the relationship between EN NH_4^+-N and overlying water inorganic nitrogen was the most prominent.

Specifically, NH_4^+-N in EN showed positive correlations with NH_4^+-N, NO_3^--N, DIN, and DTN in overlying water, among which the correlations with overlying water NH_4^+-N, DIN, and DTN were stronger, and a relatively consistent positive correlation trend was also observed with overlying water NO_3^--N. This indicates a relatively close correspondence between exchangeable NH_4^+-N in surface sediments and inorganic nitrogen levels in overlying water. In contrast, the correlations between NO_3^--N in EN and the inorganic nitrogen indicators in overlying water were generally weaker, suggesting that the consistency between exchangeable NO_3^--N in surface sediments and inorganic nitrogen in overlying water was relatively less evident.

Overall, NH_4^+-N in PMN and NO_3^--N in PMN also showed predominantly positive correlations with inorganic nitrogen indicators in overlying water, but the correlation strengths were generally lower than those of NH_4^+-N in EN. Among them, NO_3^--N in PMN exhibited certain positive correlations with NO_3^--N, DIN, and DTN in overlying water, while the correlations between NH_4^+-N in PMN and the overlying water indicators were generally weak. Overall, the different labile nitrogen fractions in surface sediments and inorganic nitrogen in overlying water were not synchronized, among which NH_4^+-N in EN showed the most pronounced response to changes in overlying water inorganic nitrogen, while the correspondences of the other fractions were relatively weak.

The above results indicate that among the labile nitrogen fractions in the surface sediments of Qilu Lake, the coupling strengths between different fractions and inorganic nitrogen in overlying water differed significantly. Specifically, exchangeable NH_4^+-N showed a closer relationship with overlying water inorganic nitrogen, suggesting that it has more direct environmental indicator significance in the nitrogen migration and transformation processes at the sediment–water interface. This finding is generally consistent with previous studies showing that bioavailable nitrogen forms in lake sediments can significantly affect inorganic nitrogen levels in overlying water[18]. Previous studies have also shown that inorganic nitrogen accounts for a relatively low proportion of total nitrogen in sediments of deep plateau lakes and temperate lakes, while organic nitrogen pools dominate, and the composition of sediment nitrogen forms and their relationships with water nitrogen indicators vary markedly among different lakes[16,17]. Meanwhile, NH_4^+-N has a stronger mobility at the sediment–water interface and is more readily released into overlying water through diffusion, resuspension, and interfacial exchange, whereas NO_3^--N is more susceptible to nitrification, denitrification, and changes in redox conditions, which often result in different correspondence patterns between these two forms and overlying water inorganic nitrogen[19]. From the results of this study, among the labile nitrogen fractions in surface sediments of Qilu Lake, exchangeable NH_4^+-N responded more sensitively to changes in overlying water inorganic nitrogen.

ISSN: 2710-0723

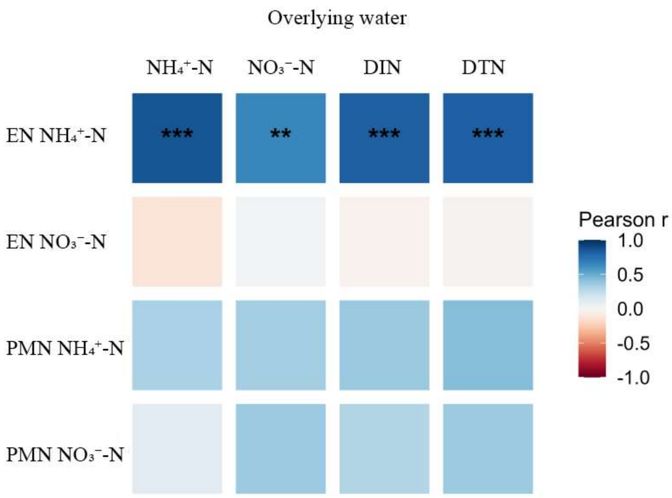

Fig. 5 Correlation heatmap between active nitrogen fractions in surface sediments and inorganic nitrogen in the overlying water of Lake Qiluhu

4.2. Driving Factors of Spatial Distribution of Nitrogen Forms in Sediments of Qilu Lake

The spatial distributions of different nitrogen fractions in the surface sediments of Qilu Lake were not synchronized, indicating that the sources, accumulation, and transformation processes of different nitrogen pools within the lake differed markedly. Overall, TN and SN exhibited more pronounced enrichment in localized areas, while different inorganic nitrogen forms in EN and PMN showed greater spatial variability, reflecting that the labile nitrogen pool was more sensitive to local environmental changes.

Among the labile inorganic nitrogen fractions, high values of NH_4^+-N in EN were mainly concentrated in localized areas on the western side of the lake, indicating relatively obvious accumulation of exchangeable NH_4^+-N in surface sediments in this area. Previous studies have shown that ammonium nitrogen in sediments generally exhibits high lability in high-nutrient lakes and may undergo strong adsorption–desorption processes at the sediment–water interface[20,21]. The western side of Qilu Lake is adjacent to inflowing rivers and village distribution areas, and is subject to considerable agricultural non-point source inputs and local domestic pollution, which may be an important reason for the relatively high enrichment of labile NH_4^+-N in this area. In contrast, high-value areas of NO_3^--N in EN were mainly located in the northeastern and north-central parts of the lake, indicating that exchangeable NO_3^--N exhibited more pronounced site-specific differences within the lake.

For PMN, both NH_4^+-N and NO_3^--N in PMN exhibited considerable spatial variation, but their high-value areas were not consistent, indicating that the composition of potential mineralization products in surface sediments differed among lake zones. Since PMN represents the labile nitrogen pool with high bioavailability in sediments, its spatial distribution is jointly affected by organic matter sources, sedimentary environment, and microbial transformation processes, and therefore tends to show interleaved high-value areas.

Regarding the total nitrogen pool structure, the spatial distributions of TN, PMON, and SN also differed. Among them, TN and SN formed relatively pronounced high values in localized areas, indicating that the accumulation of the stable nitrogen pool was more pronounced in some lake zones; while PMON was relatively high in the western side and parts of the central lake area, suggesting that these areas still had relatively high reserves of potential mineralizable organic

ISSN: 2710-0723

nitrogen. Yin et al.[11] showed that TN in the surface sediments of Qilu Lake exhibited obvious spatial differences across the lake, with relatively high nutrient pollution levels. On this basis, the present study further revealed that, in addition to TN, different labile nitrogen fractions and organic nitrogen pools also exhibited inconsistent spatial distribution patterns, indicating that the occurrence and potential transformation risks of nitrogen in surface sediments of Qilu Lake had significant spatial heterogeneity.

4.3. Mechanisms of Vertical Distribution of Nitrogen Forms in Sediments of Qilu Lake

Sediment cores can not only record the long-term evolution of lake water environment, but also reflect the combined effects of external inputs, depositional accumulation, and internal transformation on the distribution of nitrogen forms at different historical stages. Overall, the vertical distributions of nitrogen forms in the different cores from Qilu Lake exhibited obvious differences, but most sites still showed the common feature of relatively high labile nitrogen in the surface layer and relatively lower concentrations in the deep layers.

NH_4^+-N at most sites showed higher concentrations in the shallow layer than in the middle and deep layers, indicating that labile ammonium nitrogen in surface sediments is relatively more prone to accumulation or renewal. Unlike the other sites, NH_4^+-N at Q9 increased markedly with depth, with deep-layer concentrations significantly higher than those in the shallow layer, exhibiting obvious abnormality. Combined with previous studies on the effects of dredging on the storage patterns of nitrogen and phosphorus in sediment cores from the Caohai area of Dianchi Lake, it is known that dredging can alter the distribution of sediment nutrients in the short term, but as new sediments continuously accumulate, the vertical structure of nitrogen and phosphorus in the cores may be readjusted and form new distribution characteristics[22]. Therefore, the abnormally high NH_4^+-N in the deep layers at Q9 may be related to the historical disturbance background and the process of sediment redeposition in this area.

The vertical variation of NO_3^--N across different sites is generally more fluctuating than that of NH_4^+-N, particularly at Q10, Q14, and Q15 where it is more pronounced. This indicates that the distribution of NO_3^--N in sediment cores is more strongly influenced by changes in environmental conditions. Since NO_3^--N is simultaneously controlled by nitrification, denitrification, and redox conditions at the interface, its vertical pattern in sediments is generally less stable than that of NH_4^+-N[19]. In the present study, NO_3^--N in the shallow layers at some sites is relatively high, while the deep layers show alternating local increases and decreases, which also reflects differences in sedimentary environment and redox conditions among lake zones.

PMN decreased from shallow to deep layers at most sites, indicating that PMN was mainly enriched in the surface layers of the cores. Relatively fresh organic matter input and relatively high microbial activity in surface sediments made them more likely to form a higher PMN pool, while labile components in deeper sediments were continuously consumed, resulting in relatively low PMN content. Local fluctuations occurred at Q15 in the 0–4 cm interval, suggesting that the surface sedimentary environment in this area may be relatively complex.

TN remained relatively stable with depth at most sites, with only Q15 showing notable inter-layer differences, indicating that a considerable portion of nitrogen in the sediment cores of Qilu Lake had entered a relatively stable storage state, while some local sites were still influenced by strong external inputs or changes in sedimentary environment. As shown in Fig. 6, SN dominated the nitrogen pool composition in most cores, further indicating that stable nitrogen remained the primary form of nitrogen storage in Qilu Lake sediments. In contrast, PMON and EN exhibited more pronounced fluctuations across different lake zones and depth intervals, suggesting that the labile organic nitrogen pool and the currently labile inorganic nitrogen pool had stronger spatial heterogeneity.

ISSN: 2710-0723

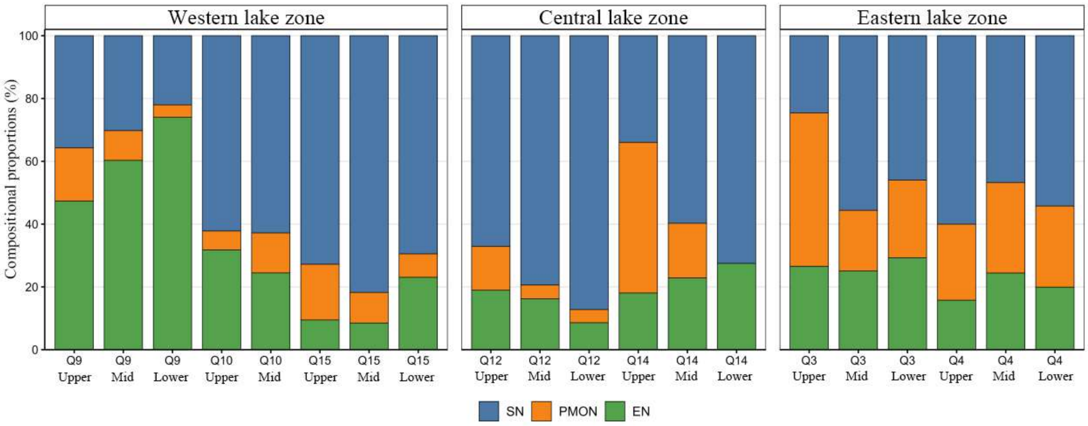

Fig. 6 Composition of nitrogen pools in core sediments across different lake zones of Lake Qiluhu

5. Conclusion

In the overlying water of Qilu Lake, nitrogen was dominated by inorganic nitrogen. NH_4^+-N concentrations were generally low, with most sites below 1 mg·L^{-1}, while NO_3^--N concentrations were relatively high. The spatial variations of DIN, DTN, and TN were generally consistent with that of NO_3^--N, and DIN predominated in DTN. At Site Q9, NH_4^+-N, NO_3^--N, DIN, DTN, and TN in the overlying water all reached relatively high levels, making it one of the most prominent sites for nitrogen enrichment within the lake.

The different nitrogen fractions in surface sediments of Qilu Lake exhibited significant spatial heterogeneity. The high-value areas of TN, PMON, SN, and various forms of EN and PMN did not completely overlap within the lake, indicating that the occurrence patterns of different nitrogen pools in surface sediments were not synchronized. Overall, TN and SN exhibited more pronounced enrichment in localized areas, while different inorganic nitrogen forms in EN and PMN showed greater spatial variability.

The release contributions of nitrogen forms in sediment cores differed significantly among lake zones. The differences among sites in the western zone were the most pronounced, with Q9 characterized by a relatively high contribution of NH_4^+-N release, and Q15 dominated by PMN release contribution. In the central zone, PMN contributions were dominant, but the relative contributions of NH_4^+-N in EN and NO_3^--N in EN still differed between sites. In the eastern zone, the overall release contribution structure was relatively stable, with PMN consistently dominating.

The vertical distribution characteristics of different nitrogen fractions in sediment cores showed obvious differences. At most sites, NH_4^+-N was relatively high in the shallow layers, while at Q9 it increased markedly with depth. NO_3^--N fluctuated considerably among sites, with Q10, Q14, and Q15 showing more pronounced variations. PMN generally decreased from shallow to deep layers. TN remained relatively stable with depth at most sites, with only Q15 showing notable changes. Overall, the vertical distributions of nitrogen forms in sediment cores of Qilu Lake exhibited both common features and obvious differences among lake zones.

The relationships between labile nitrogen fractions in surface sediments and inorganic nitrogen in overlying water are differentiated. Correlation analysis showed that NH_4^+-N in EN had the most significant correlations with NH_4^+-N, DIN, and DTN in overlying water, while the correlations of NO_3^--N in EN and the two inorganic nitrogen fractions in PMN with inorganic nitrogen in overlying water were relatively weak. This indicates that exchangeable NH_4^+-N in the labile nitrogen pool of surface sediments in Qilu Lake is more responsive to changes in inorganic nitrogen in overlying water.

ISSN: 2710-0723

Acknowledgments

This work was funded by National Major Science and Technology Program for Jing-Jin-Ji Regional Integrated Environmental Improvement-National Science and Technology Major Project (2026ZD1214901), Water Pollution Control and Treatment (No.2012ZX07102-004), National Key R & D Program of China (No.2017YFA0605202-001), Ministry of Science and Technology of the People's Republic of China (No.2015FY110900-05) and National Natural Science Foundation of China (No.41103070).

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