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Research article

### Patterns of internal nitrogen and phosphorus loadings in a cascade reservoir with a large water level gradient: Effects of reservoir operation and water depth

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#### ABSTRACT

Internal nutrient loadings pose a high risk of being an additional N and P source, exacerbating eutrophication and deteriorating water quality. In this study, we selected the Daheiting Reservoir (DHTR) in North China, with a pronounced water level gradient, to investigate internal N and P loadings, estimate N and P fluxes across the sediment-water interface based on the pore water profiles, and reveal the potential effects of water discharge from an upstream reservoir and high-intensity cage aquaculture on the risks of internal N and P release. The results indicated that DHTR presented with severe internal nutrient loadings, and the N and P fluxes showed significant spatiotemporal variations.  $NH_4^+ - N$  and soluble reactive phosphorus (SRP) fluxes were higher in deep areas (averages of 26.14 and 9.9 mg m<sup>-2</sup>  $d^{-1}$ , respectively) than in shallow areas near inflows (averages of 5.0 and 1.24 mg m<sup>-2</sup>  $d^{-1}$ , respectively). Unexpectedly, the estimated  $NH_4^+ - N$  and SRP fluxes were the lowest in summer (averages of 3.94 and 0.33 mg m<sup>-2</sup>  $d^{-1}$ , respectively), which may have been influenced by seasonal thermal stratification and copious discharge from the hypolimnion of the upstream reservoir (Panjiakou Reservoir). Comparison of annual internal and external N and P loadings revealed that water discharge from the upstream Panjiakou Reservoir was the dominant source of N and P to the reservoir, contributing up to 83.6% of N input and 55.4% of P input. The internal P loading also contributed to water eutrophication to a great extent, accounting for 34.7% of the total P input. Our results highlight the impact of upstream reservoir discharge operation on downstream reservoir water quality and the importance of controlling the internal nutrient loading in cascade reservoirs, and further provide theoretical and practical foundations for the development of policies and strategies to conserve reservoir ecosystems.

#### 1. Introduction

Since the 20th century, various degrees of eutrophication have been reported in many lakes and reservoirs worldwide (Conley et al., 2009; Paerl et al., 2018; Zhang et al., 2020). Nitrogen (N) and phosphorus (P) are critical nutrients for maintaining primary productivity in lake and reservoir ecosystems, and are also the major limiting elements for eutrophication (Elser et al., 2007; Schindler et al., 2016; Seitzinger and Kroeze, 1998). Intense increases in N and P in the water column can aggravate toxic cyanobacterial blooms, posing a threat to the water

supply of freshwater ecosystems (Orihel et al., 2013; Ye et al., 2009). To reverse lake and reservoir eutrophication, considerable efforts have been made to reduce external N and P pollution. However, for water bodies with high internal loading, even if external nutrients inputs are effectively controlled, the continued release of N and P from sediments could continue to hinder water quality improvement for decades, delaying the recovery process (Jeppesen et al., 2005; Sugimoto et al., 2014; Wu et al., 2017). Therefore, management and reduction of the internal N and P loadings are vital for effective freshwater ecosystem restoration.

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In the process of exploring the mechanism of lake and reservoir eutrophication, it is crucial to analyze the nutrient migration mechanism across the sediment-water interface (SWI) (Lei et al., 2018). The nutrient transportation and conversion process across the SWI is complex, and may be influenced by environmental and internal factors. The main environmental factors are temperature, pH, dissolved oxygen (DO), redox potential, and hydrodynamic disturbance; internal factors mainly include bioturbation, and nutrient forms in sediment and their concentration (Sondergaard et al., 2003; Song et al., 2017; Yang et al., 2020; Zhang et al., 2008). For example,  $NH_4^+ - N$  is generated by the mineralization of organic N in sediment and adsorbed on the mineral surface or stored in pore water in the form of  $NH_4^+ - N$ ;  $NH_4^+ - N$  in the pore water is then released upwards and becomes an important source of N in the overlying water (Markou et al., 2007; Wen et al., 2018). Labile-P is regarded as immediately available P, combined with sediment particles through physical adsorption, which is unstable and readily released into overlying water and promotes eutrophication by direct algal uptake (Rydin, 2000; Zhou et al., 2001).

The release of nutrients in shallow lakes and reservoirs is often affected by sediment resuspension (Havens et al., 2007; Tammeorg et al., 2015). Strong resuspension may result in a higher nutrient concentration gradient at the SWI than before the resuspension event, whereas mild resuspension may in turn dilute the nutrient concentration of the first few millimeters of surface sediment (Tammeorg et al., 2020). Nevertheless, other mechanisms may also play important roles in N and P release from sediment. For example, N and P release increases as the temperature increases, indicating that temperature is a key driver in shallow ecosystems (Jiang et al., 2008; Yang et al., 2020). Furthermore, extensive studies have proved that reductive dissolution of Fe-bound P was the major mechanism of P mobilization in sediments (Ding et al., 2016; Markovic et al., 2019). For sub-deep lakes and reservoirs with obvious water level gradients, shallow regions (<10 m) near inflows may lack thermal stratification throughout the year (Chen et al., 2018), and the main N and P release mechanisms may be similar to those of shallow lakes. However, in deep water areas (<50 m), seasonal stratification can be the primary factor affecting physical, chemical, and biological processes and nutrient circulation (Chen et al., 2018; Wang et al., 2016). Longitudinal development of thermal stratification can lead to variation in DO in the water column, thereby affecting nutrient release potential from sediment (Nowlin et al., 2005). Accelerated sediment P release during seasonal anoxia was found in stratified lakes and reservoirs (Anderson et al., 2021; Wang et al., 2016). Seasonal exchange and sudden convective mixing between the bottom and upper water may break the temporary stratification and allow nutrients to flow into the upper water, resulting in water quality deterioration and algal blooms. This is unlikely to occur in deep lakes and reservoirs where the water is sufficiently deep to allow permanent thermal stratification (Chen et al., 2018). Therefore, more attention should be paid to the risk of internal N and P release of sub-deep lakes and reservoirs.

There are many cascade reservoirs in the world, especially in developing countries, and are used for hydropower, water supply, flood control, and other purposes (Moran et al., 2018). The accumulation of nutrients in artificial reservoirs and the high risk of sediment nutrient release have been well documented worldwide (Chen et al., 2019; Lima et al., 2022; Rocha and Lima et al., 2022). River damming can retain nutrients, accelerate the eutrophication process of a reservoir, and affect nutrient bioavailability in downstream reservoirs (Maavara et al., 2015; Chen et al., 2020). Additionally, the management and operation of upstream reservoirs can affect downstream reservoirs and rivers (Chen et al., 2018). For example, water discharged from an upstream reservoir would influence the water temperature of a downstream reservoir and disrupt seasonal stratification, thereby affecting the nutrient biogeochemical cycle in aquatic ecosystems and the internal loadings of the reservoir (Olsson et al., 2022). To date, there have been few studies on the impact of upstream reservoir drainage operation on the internal

nutrient loading.

Daheiting Reservoir (DHTR) is the key node of the cascade reservoir in the water diversion project from Luanhe River to Tianjin City in North China. The water flows from the Panjiakou Reservoir through Luanhe River into DHTR and thereafter into the Yuqiao Reservoir via Li River (Wen et al., 2020). Due to the disorderly development of cage aquaculture in DHTR since the 1990s, the accumulation of fish baits and excrements have rapidly deteriorated the water body and accelerated eutrophication (Wen et al., 2019b). To improve the water quality, the regional government mandated fish cage cleaning between October 2016 and May 2017. However, due to the long-term contamination of DHTR caused by cage culture, N and P have been continuously released from the sediment into the water column, resulting in the frequent occurrence of large cyanobacteria bloom in recent years (Liu et al., 2020; Wen et al., 2020). In China, many water bodies have been polluted severely by aquaculture (Jia et al., 2017; Meng et al., 2021; Wang et al., 2021). However, the spatiotemporal variations of internal N and P fluxes following high-load accumulation of nutrients have rarely been studied.

We hypothesized that the N and P cycling mechanisms may differ between upstream and downstream for reservoirs characterized by a pronounced water level gradient. N and P cycling mechanisms in the shallow areas near inflows may resemble in shallow lakes, while the mechanisms may be influenced by seasonal thermal stratification in the deep areas near the dam. We further hypothesized that regulatory operation of the upstream reservoir could affect the redox environment at the SWI of the downstream reservoir, thus affecting the risk of N and P release. In addition, the N and P release rates may be comparatively higher in regions with abundant fish cages as a result of high endogenous pollution loadings. The aims of this study were to: (1) Investigate the spatiotemporal variations in the physicochemical properties of the overlying water and the internal N and P loadings of the reservoir with pronounced water level gradients; (2) reveal the potential impact of water discharge from the upstream reservoir on the spatiotemporal heterogeneity of N and P release from the sediment; and (3) evaluate the internal N and P loading intensities of the reservoir affected by intensive aquaculture and the relative impacts of external and internal loadings. The results of this study will provide a reference for eutrophication management in cascade reservoirs.

#### 2. Materials and methods

#### 2.1. Study area

DHTR (40°12′-40°21′N, 118°15′-118°19′E) is located on the main stream of the Luanhe River, 30 km downstream of Panjiakou Reservoir (PJKR) in Hebei Province, China. It has a temperate monsoon climate, with warm and wet summers and cold and dry winters. DHTR was built and activated in 1986, controlling a watershed area of 35,100 km<sup>2</sup>, with a total length of 22 km and a total storage capacity of  $337 \times 10^6$  m<sup>3</sup> (Wen et al., 2019b). DHTR is the key intermediate reservoir in the water diversion project from Luanhe River to Tianjin City, with the main function of meeting the water demand of Tianjin and Tangshan cities. DHTR receives regulated water from PJKR, raises the water level, and transfers the water to Yuqiao Reservoir (YQR) (Gong et al., 2019; Wen et al., 2020). Over the past 20 years, fish cages have inundated the reservoir and were distributed in the areas covering the sampling sites denoted as D1-D5 in the present study (Fig. 1), resulting in the accumulation of high nutrient and organic matter contents in the sediments. The external sources of DHTR include the discharge from the upstream PJKR, inflows from Sahe River (a tributary of Luan River), and non-point sources from surface runoff surrounding the reservoir. The annual rainfall distribution in DHTR was not uniform, and rainfall was mainly concentrated in July and August (Fig. 2a). From April-June, discharge from PJKR was moderate, increased sharply due to the influence of heavy rain during July and August, and then decreased to an extremely low level in September and the subsequent months (Fig. 2b). Meanwhile,

the manager of the cascade reservoirs wishes to decrease the water temperature of DHTR by increasing the discharge from PJKR in summer, so as to curb the outbreak of harmful algal blooms.

#### 2.2. Sample collection and processing

Sampling was performed in March (winter) (just after the icebreaking), May (spring), August (summer), and November (autumn) (ice-free period) in 2018. To understand the variations of water quality in the reservoir, 35 sampling sites were uniformly distributed (Fig. 1) to collect the overlying water samples using polyethylene bottles. Surface sediment of the 35 sites were sampled in the initial sampling campaign (March 2018) to investigate sediment nutrient loadings. To examine the variation of water temperature (WT) and DO stratification in DHTR, a free flow depth-setting sampler (HYDRO-BIOS KIEL, Germany) was used to sample the water at a 5-m interval in front of the dam (D1 site). WT, DO, pH, and oxidation-reduction potential (ORP) of water samples were measured in the field using a multi-parameter water quality meter (YSI, Yellow Springs, OH, USA). Water depth (WD) was measured with a portable ultrasonic water depth meter (SM-5, Laylin, USA). Seven representative sites were selected from the 35 sampling sites for sediment core collection with a gravity corer (D = 90 mm; L = 500 mm). D1–D3 sites (average water depth ≥14.6 m) were located in deeper

water areas near the dam, and D4–D7 sites (average water depth  $\leq$ 11 m) were located in the relatively shallow areas near the inflows (Fig. 1).

After sampling, the sediment cores were immediately transported to the laboratory with care taken to avoid disturbing the SWI. Sediment cores were not collected at D7 site during the summer as the WD was too low to permit sampling boat access. All cores were incubated in a constant-temperature water tank in the dark. The incubation temperature was similar to in-situ water temperature ( $\pm 2$  °C). The high-resolution peeper (HR-peeper) was vertically inserted into the cores to sample sediment pore water and was retrieved 2 d after deployment. Then, the pore water was sampled to a 96-well microplate for ammonium ( $NH_4^+ - N$ ) and soluble reactive phosphorous (SRP) concentration measurements. The HR-peeper and the sampling operation were described in detail in Xu et al. (2012) and Johnston et al. (2009).

#### 2.3. Calculation of N and P diffusive fluxes

The vertical profiles of  $NH_4^+ - N$  and SRP in pore water were used to estimate the diffusion fluxes across the SWI according to Fick's first law of diffusion (Ullman and Aller, 1982), as follows:

$$F = \varphi D_s \frac{\partial c}{\partial x}\Big|_{x=0}$$



Fig. 1. Locations of sampling sites and map of annual average water depths at Daheiting Reservoir. The areas from the red solid line to the downstream dam were the areas where the fish cages were distributed. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)



Fig. 2. (a) Precipitation and (b) inflows and outflows of Daheiting Reservoir in 2018 (Data source: Haihe River Water Conservancy Commission, Ministry of Water Resources of China).

where *F* represents the  $NH_4^+ - N$  and SRP diffusive fluxes ( $mg m^{-2} d^{-1}$ ),  $\varphi$  is the surface sediment porosity,  $\frac{\partial c}{\partial x}|_{x=0}$  represents the  $NH_4^+ - N$  and SRP concentration gradients between the surface sediment and the bottom of overlying water, and  $D_s$  represents the sediment diffusion coefficients of  $NH_4^+ - N$  and SRP ( $10^{-6} cm^{-2} s^{-1}$ ), which could be calculated according to empirical equations provided by Ullman and Aller (1982), as follows:

$$D_s = \varphi D_0 \dots \varphi < 0.7$$

$$D_s = \varphi^2 D_0 \dots \varphi < 0.7$$

where  $D_0$  is the theoretical diffusion coefficient for infinite dilution of solution.

#### 2.4. Laboratory analysis

The DO concentration at SWI and the oxygen penetration depth (OPD) of the sediment cores were measured using a Unisense Microprofiling System (Unisense A/S, Aarhus, Denmark). Porosity was measured with a cutting ring (Graca et al., 2004). Loss on ignition (LOI) was determined by calcinating the sediment at 550 °C for 4 h. Fresh sediment was freeze-dried under vacuum conditions and then ground and passed through a 100-mesh sieve for analysis. Total organic carbon (TOC) was determined by potassium dichromate oxidation-ferrous sulphate titrimetry (Jin and Tu, 1990). Total phosphorous (TP) contents in sediment were analyzed by molybdenum blue method to determine the phosphate concentration in all extracts after sample calcination at 450 °C and acid extraction (Pardo et al., 2003). Sediment total nitrogen (TN) was extracted with alkaline potassium persulfate and evaluated spectrophotometrically. Labile phosphorus (labile-P) in the sediment was extracted with 1  $mol L^{-1} NH_4Cl$  and analyzed by UV-VIS spectrophotometry (SHIMADZU UV-2550, Japan). Labile nitrogen ( $NH_4^+ - N$ ,  $NO_3^- - N$ , and  $NO_2^- - N$ ) in the sediment was extracted with 2 mol  $L^{-1}$ KCl and the extracts were centrifuged, filtered, and measured (Shannon et al., 2011). The  $NH_4^+ - N$ ,  $NO_3^- - N$ , and  $NO_2^- - N$  concentrations in the water samples and extracts were determined with Nessler's reagent, UV spectrophotometry, and N-(1-Naphthyl) ethylenediamine dihydrochloride spectrophotometry, respectively. SRP was determined by ammonium molybdate spectrophotometry (Murphy and Riley, 1962). The TN and TP in water columns were determined by digestion of alkaline potassium persulfate at high temperature (Jin and Tu, 1990).

#### 2.5. Statistical analysis

Significant differences between samples were analyzed by the nonparametric Kruskal–Wallis test. Spearman's rank correlation was used to test the correlations between variables. Statistical analyses were performed using SPSS Statistics v. 22.0 (IBM Corp, Armonk, NY, USA). Data were plotted with Origin 2021b (OriginLab Inc, Northampton, MA, USA) and Arcmap v. 10.2 (ESRI Inc, Redlands, CA, USA). Kriging interpolation in Arcmap was used to display spatial variations in the physicochemical properties of water columns and sediments.

#### 3. Results

#### 3.1. Basic physicochemical properties of overlying water and sediment

The physicochemical properties of the overlying water in DHTR varied spatiotemporally (Table S1). The pH varied from 7.16 to 10.07, indicating an alkaline water.  $NO_3^- - N$  was the predominant species of DIN in the overlying water. The SRP concentration was higher in spring and autumn than in winter and summer. The TN/TP value varied from 26.7 to 356.9 in DHTR. WD increased in the direction from the reservoir entrance to the dam (Fig. 1). During spring, summer, and autumn, WT was lower in the inlet areas than the main reservoir and the areas near the dam, whereas the distribution pattern was reversed in winter (Fig. S1).

The spatial patterns of nutrients and organic matter in the surface sediments of DHTR are shown in Fig. 3. Sediment TN,  $NO_3^- - N$ ,  $NH_4^+ - N$ , TOC, and LOI all gradually decreased from the deep areas near the dam to the reservoir entrance. The labile-P content was slightly high in the largely partial deep areas (Fig. 3). As shown in Fig. 4, the OPDs were deeper in spring and summer than in autumn and winter. Spatially, the OPDs were deeper and the DO concentrations were higher in the shallow water areas, whereas hypoxia was more intense in the deep areas.

## 3.2. Vertical variations in water temperature and DO and their seasonal stratification

Vertical variations in WT and DO in deep regions near the DHTR dam (D1 site) are shown in Fig. S2. DHTR had obvious seasonal water temperature stratification. Spring and summer were the thermal stratification periods. With the decrease of temperature, thermal stratification receded in autumn and disappeared altogether in winter. Due to the lack of oxygen exchange in the vertical direction affected by thermal stratification, DO stratification was also obviously developed. DO concentration was lower in hypolimnion than in surface water in winter. In summer, the DO concentration decreased whereas DO stratification was weak (Fig. S2). This was because the water bodies were partially vertically mixed under the influence of rainfall and large flow discharge scheduling during July and August (Fig. 2), resulting in DO in hypolimnion being partially replenished (5.84  $mg L^{-1}$ ), and DO in surface



Fig. 3. Spatial patterns of nutrients and organic matter in the surface sediments.



Fig. 4. DO profiles across the SWI from seven sampling sites (D1-D7) in four seasons. The 0-mm depth represents the SWI.

water being depleted (8.55  $mgL^{-1}$ ). After the discharge dispatching ceased in September and October (Fig. 2b), DO stratification was especially pronounced in autumn and hypoxia was more severe.

## 3.3. Vertical distributions of $NH_4^+ - N$ and SRP in sediment-water profiles

The average  $NH_4^+ - N$  concentrations in pore water were 6.65

 $mg L^{-1}$ , 8.37  $mg L^{-1}$ , 9.77  $mg L^{-1}$ , and 8.13  $mg L^{-1}$  in winter, spring, summer, and autumn, respectively (Fig. 5). The average  $NH_4^+ - N$  concentration was highest in summer and lowest in winter. Spatially, the  $NH_4^+ - N$  concentration was higher in the deep areas (D1–D3) than the relatively shallow areas (D4–D7) in winter, spring, and autumn. The  $NH_4^+ - N$  concentration showed little change above the SWI; whereas, it generally increased with depth below the SWI. In the relatively shallow areas (D4–D7), the  $NH_4^+ - N$  concentration showed strong vertical

variation in summer and an extremely small change in winter. However, the  $NH_4^+ - N$  profiles in the deep areas (D1–D3) showed significantly weak variations in summer.

The SRP concentration in pore water showed an evident spatiotemporal variation (Fig. 6). The average SRP concentrations in pore water were 1.58 mg  $L^{-1}$ , 1.82 mg  $L^{-1}$ , 0.39 mg  $L^{-1}$ , and 0.65 mg  $L^{-1}$  in winter, spring, summer, and autumn respectively, which indicated that SRP concentration was significantly higher in winter and spring than in autumn and summer (p < 0.05). Compared to the shallow areas near the inflows (D4–D7), the deeper areas near the dam (D1–D3) had higher SRP concentrations and stronger vertical variations. Different from the variation of  $NH_4^+ - N$  profiles, the SRP concentration increased steadily, peaked at 10–30 mm, and gradually decreased thereafter. The vertical distribution of SRP changed dramatically in spring and winter. Interestingly, the vertical variations of SRP were minimized in summer and were basically unchanged.

#### 3.4. $NH_4^+$ -N and SRP fluxes at the SWI

The  $NH_4^+ - N$  fluxes showed spatiotemporal variability with a range of 0.39–56.80 mg m<sup>-2</sup> d<sup>-1</sup>; the average values were 12.08 ± 1.43, 16.23 ± 2.73, 3.94 ± 1.03, and 24.33 ± 2.16 mg m<sup>-2</sup> d<sup>-1</sup> in winter, spring, summer, and autumn, respectively (Fig. 7). In summer, the  $NH_4^+ - N$  flux reached the lowest level and was significantly lower than that in autumn (p < 0.05) (Fig. 7). The  $NH_4^+ - N$  flux gradually increased from the shallow water areas near the entrance to the deep areas (Fig. 8). Spearman's correlation analysis showed that the  $NH_4^+ - N$  flux had no correlation with the N forms in the overlying water (Table S4).

The SRP fluxes varied from 0.01 to 24.79  $mg m^{-2} d^{-1}$ , reaching the lowest level in summer (average  $0.33 \pm 0.14 mg m^{-2} d^{-1}$ ) and the highest level in winter (average  $13.12 \pm 2.74 mg m^{-2} d^{-1}$  (Fig. 7). Spatial variation in SRP flux showed a similar pattern to that of  $NH_4^+ - N$  flux (Fig. 8). No correlation was found between SRP flux and TP or SRP in the overlying water (p < 0.05) (Table S4).

#### 4. Discussion

#### 4.1. Conditions of water column and sediment contamination

According to the Chinese environmental quality standard for surface water (GB3838-2002), the TN concentration in DHTR exceeded class five (V) throughout the year, especially in winter. The TP concentration was in class three (III) during spring and autumn and class four (IV) during winter and summer. Guildford and Hecky (2000) suggested that the TN/TP value was indicative of which nutrient could become limitation for growth in lakes and oceans. N-deficient growth was apparent at TN/TP < 9, whereas P-deficient growth occurred when TN/TP > 22.6, and at intermediate ratios, either N or P can become deficient. In this study, we observed P limitation throughout the year.

Based on the assessment criteria for TN and TP pollution in sediments developed by the U.S. Environmental Protection Agency (EPA) (USEPA, 2002), the contents of both reached severe pollution levels in this study. Organic matter in sediment is one of the important indicators of endogenous pollution and organic nutrient levels in lakes and reservoirs, which plays an essential role in the N and P migration and release process (Zhao et al., 2013). Sediment TN had a significant positive correlation with TOC (p < 0.01) (Table S3), suggesting that sediment TN was mainly derived from organic matter. By contrast, sediment TP was not correlated with TOC, which indicated that TP was determined by other sediment constituents than organic matter (Knosche, 2006). Due to the different stability of N release and transformation in different organic matter types, the C/N ratio is commonly used to reveal the organic matter source and type (Thornton and Mcmanus, 1994). Studies have shown that higher C/N ratios generally indicate terrigenous organic matter, whereas lower C/N ratios represent organic matter mainly from the water bodies (Qiu et al., 2016; Wang et al., 2014). The surface sediment C/N ratio of DHTR was within a relatively low range (4.9-15) with a mean value of 8. This result showed that the organic matter in DHTR surface sediments was mainly from the water bodies, i. e., from the oxidation decomposition of zooplankton, phytoplankton,



**Fig. 5.** Variations in the  $NH_4^+ - N$  concentration measured by HR-peeper in overlying water and sediment profiles from seven sampling sites (D1–D7) in Daheiting Reservoir. The 0-mm depth represents the SWI.



Fig. 6. Variations in the SRP concentration measured by HR-peeper in overlying water and sediment profiles from seven sampling sites (D1–D7) in Daheiting Reservoir. The 0-mm depth represents the SWI.



Fig. 7. NH $_{\rm +}^{+}$ -N and SRP fluxes across the SWI in different seasons. The same lowercase letters indicate no significant differences, and different lowercase letters indicate significant differences (p < 0.05).



**Fig. 8.** Diffusive fluxes of  $NH_4^+ - N$  and SRP at the SWI from the seven sampling sites (D1–D7) in Daheiting Reservoir.

and fish baits and excrements. The higher TOC, TN,  $NH_4^+ - N$ , and  $NO_3^- - N$  contents in the areas where the fish cages were distributed further indicated that the internal pollution of DHTR was mainly caused by human activities, especially the fish cage culture. Dam construction decreases water flow velocity and increases sediment residence time, which consequently allows the deposition of organic matter in large quantities in the reservoir areas, especially in the stagnant areas near the dam (Bing et al., 2016; Fremion et al., 2016). Therefore, the effect of the dam on retention and accumulation of organic matter was also an important reason for the high nitrogen and organic matter contents in the deep areas near the dam of DHTR.

#### 4.2. Patterns of $NH_4^+ - N$ and SRP in pore water

Sediment pore water acts as the medium of nutrient exchange at the SWI (Lei et al., 2018). The diffusion intensity of sediment nutrients mainly depends on their concentration gradients in pore water (Lerat et al., 1990; Wen et al., 2019a). The results showed a difference in the  $NH_4^+$  – N concentration between the overlying water and sediment pore water (Fig. 5). Below the SWI, the  $NH_4^+ - N$  concentration increased with depth, showing the migration and diffusion capacity due to the  $NH_4^+ - N$  concentration gradient in sediment pore water, and reflecting the upward migration process of  $NH_4^+ - N$  across the SWI, which was one of the sources of N in water columns (Wang et al., 2021). The DO concentration in sediments decreased continually with depth until it reached 0  $\mu$ mol L<sup>-1</sup> (Fig. 4). This means that the anaerobic degree in sediments continues to deepen, aerobic bacteria are inhibited, and anaerobic bacteria predominate, resulting in nitrification diminishing while denitrification and ammonification are augmented in the deep sediment layers, which is conducive to  $NH_4^+ - N$  accumulation (Alvarez and Vogel, 1995; Yin et al., 2002). Therefore, the  $NH_{4}^{+} - N$  concentration gradually increased with sediment depth.

At the sampling sites in shallow water areas upstream, the  $NH_4^+ - N$  concentrations in pore water were the highest with the strongest vertical variation in summer, while reached the lowest in winter with little change in vertical distribution. This seasonal variation agrees with the results of prior studies on shallow eutrophic lakes and reservoirs (Wen et al., 2018; Yang et al., 2020). This may be because of the microbial activity at the sediment surface intensifying with the temperature increasing during warm seasons in the shallow water areas, resulting in organic nitrogen mineralization being promoted, and more  $NH_4^+ - N$  being produced and released into sediment pore water. However, the  $NH_4^+ - N$  concentration in sediment pore water was lower in summer than in the other seasons at D1–D3 sites near the downstream dam, which indicated that the capacity of sediment supplying  $NH_4^+ - N$  to pore water decreased in the deep areas in summer.

The overall SRP profile presented with a release peak in sediment pore water (Fig. 6), which was consistent with observations of previous studies (Wen et al., 2019a; Yang et al., 2020; Yu et al., 2017). This depth was the accumulation area of bioavailable phosphorus, which was a valuable indicator of the internal P loading process occurring recently in sediments (Ding et al., 2010). Under geological diagenesis, iron hydroxide is deoxidized in the condition of relative reduction, resulting in the dissolution and release of P bound with iron hydroxide (Markovic et al., 2019), which also explains the higher SRP concentration in pore water in the deep areas with lower oxygen concentrations at the SWI (Fig. 4). The relatively low SRP concentrations in pore water in the shallow water areas may have been directly related to the labile-P content in the sediment (Fig. 3). In addition, aquatic plants were abundant at the bottom of water columns in the partial shallow water areas of DHTR. Low SRP concentrations may also be related to nutrient uptake and assimilation by submerged plants (Wen et al., 2019b). The pore water SRP concentrations in previous studies of shallow lakes and reservoirs increased rapidly during the warm seasons under high

temperature (Wen et al., 2018; Yang et al., 2020). Interestingly, the seasonal variation in DHTR was on the contrary. The SRP concentration in pore water was highest in winter and spring and lowest in summer. Due to the discharge from the low-temperature hypolimnion of upstream deep-water reservoir in summer, the temperature in the shallow water areas upstream of DHTR was much lower than air temperature (Fig. S1), and the low temperature at the SWI in deep areas was also retained because of thermal stratification. Thus, the elevated air temperature in summer was not as predominant in promoting SRP generation as it was in shallow lakes and reservoirs. In addition, DO was supplemented at the bottom of the water column most likely due to the water convection being enhanced in DHTR under the rainstorms and large discharge scheduling in summer (Fig. 2), especially in the shallow areas, where P was oxidized and sorbed on iron oxyhydroxides, reducing the SRP concentration in pore water.

## 4.3. Nutrient fluxes across the SWI and the possible controlling mechanisms

Sediment release may be a major source of the N and P concentration increases in water columns (Conley et al., 2009; Wang et al., 2015). The results indicated that the N and P fluxes at the SWI varied spatiotemporally (Figs. 7 and 8). Compared with other trophic lakes and reservoirs worldwide, N and P fluxes were substantially higher in DHTR (Table S5) under the high sediment nutrient loadings caused by fish cage aquaculture. Ammonium migration across the SWI in lakes and reservoirs is complex and mainly affected by redox potential, microbial activity, organic matter decomposition, and N loading in sediment (Zheng et al., 2016). The average  $NH_4^+$  – N release rate across the SWI was the highest in autumn, which was 6.2 times of that in summer. Affected by rainstorms and large-flow discharge scheduling in DHTR in summer (Fig. 2), the loose surface sediments in the shallow water areas near inflows would be strongly resuspended, resulting in complete exchange of sediment pore water with the water body above the SWI. In addition, under the influence of surface runoff caused by rainfall, the Sahe River at the reservoir entrance imported abundant nutrients. Consequently, the  $NH_{4}^{+} - N$  concentration gradients across the SWI in the shallow water areas decreased, inducing the fluxes estimated by Fick's first law to be less in summer in this study. This can be proved by the higher concentration of  $NH_4^+ - N$  in the water column in the summer (Table S2). In stratified reservoirs, the temporary stratification can inhibit mass transfer between the upper and lower layers (Yang et al., 2021). In deep areas near the dam downstream, DO concentration at the SWI was at a very low level (Fig. 4), which promoted  $NH_4^+ - N$  generation and accumulation. In summer, stable thermal stratification formed in the water columns (Fig. S2). The epilimnion prevented  $NH_4^+ - N$  in the hypolimnion from diffusing into the surface water, resulting in a lower gradient at the SWI as mentioned above, which further inhibited the release potential of  $NH_4^+ - N$  in the sediment. Additionally, compared with autumn, the higher DO concentration of water column and the larger OPDs at the SWI in summer were also not conducive to the accumulation of  $NH_4^+$  – N in the pore water (Figs. S2 and 4), resulting in lower  $NH_4^+ - N$  flux at the SWI. The vertical convective diffusion of water strengthened as thermal stratification disappeared in autumn, which induced the  $NH_4^+ - N$  concentration to decrease and increase in the bottom and surface water, respectively. When the upstream reservoir stopped large drainage flows (Fig. 2b), DO stratification intensified and OPDs were minimal in autumn (Figs. S2 and 4), which demonstrated that hypoxia was obvious in surface sediment. Under such anaerobic conditions, nitrification can be inhibited and organic N mineralization could be promoted, inducing  $NH_4^+ - N$  accumulation in the surface sediment and pore water (Zhong et al., 2021). This may have been the important cause of the  $NH_4^+ - N$  flux was higher in autumn. Furthermore, correlation analysis results showed that the  $NH_4^+ - N$  flux was negatively correlated with OPD (p < 0.05) (Table S4).  $NH_4^+ - N$  in the

overlying water was not affected by diffusive flux at the SWI.  $NH_4^+ - N$  flux was weakly correlated with the nutrient contents in the overlying water and the water environmental parameters, while it was negatively correlated with TP (p < 0.05) (Table S4).

In this study, the SRP release flux was consistent with the spatiotemporal variation pattern of the SRP concentration in pore water. As shown in Fig. 6, P in the surface sediment had an upward migration trend at all sampling sites, which may have a considerable influence on the nutrient state of water columns in DHTR. The average release rate of SRP in winter was approximately 6.7 times higher than that in summer, which was inconsistent with previous research results that the risk of internal P release was far greater in summer than winter (Liu et al., 2016; Moodley et al., 1998; Yang et al., 2020; Yu et al., 2016). The relevant factors affecting sediment P release mainly include redox potential, DO, temperature, pH, and hydrological conditions (Jin et al., 2006; Kim et al., 2003). When the DO concentration is at a high level across the SWI, the redox condition of the surface sediment becomes conducive to iron oxide or iron hydroxide formation (Markovic et al., 2019). Phosphate can be efficiently sorbed to ferric compounds and hydroxide available in the sediment. However, ferric ion is reduced to ferrous ion under anaerobic conditions and subsequently both iron and sorbed phosphate return into solution (Sondergaard et al., 2003; Xu et al., 2013). In agreement herewith, some researchers have suggested that internal P release could be partially inhibited by the oxic-trapped microlayer at the SWI, while P can be freed when the oxidized microlayer is chemically reduced with the depleted DO at the SWI (Glud et al., 2007; Penn et al., 2000). During the frozen period, the layer of ice and snow could reduce light penetration and inhibit oxygen production by aquatic photosynthetic organisms, as well as oxygen diffusion across the air-water interface (Greenbank, 1945; Magnuson et al., 1985). Therefore, the oxygen consumption rate is greater than the oxygen replenishment rate, and DO decreases in part of water columns in nutrient-rich waters, which is defined as over-winter hypoxia (Tellier et al., 2022; Yang et al., 2016). In relatively deeper systems, over-winter hypoxia only develops near the sediment while the upper water retains higher DO concentrations (Kalejs and Thesis, 2017; Tellier et al., 2022). Similarly, winter samples in this study were collected at the end of the ice period (immediately after the ice-breaking), when DO was not fully supplemented in the hypolimnion. In deep areas, there was an obvious hypoxia phenomenon at the SWI (Fig. 4). Moreover, the P release rate from sediment was significantly negatively correlated with the OPD (p < 0.05) and ORP (p < 0.01) of the overlying water (Table S4). This indicated that P in the sediments may have been subject to the same release mechanism in DHTR. In addition, some studies have reported that an increase in pH can free P from its binding with ferric complexes through the competitive exchange between hydroxyl ions and the bound P ions, intensifying the potential for P release (Christophoridis and Fytianos, 2006; Kim et al., 2003). The pH of overlying water in this study was higher in winter and spring than in summer and autumn (Table S1), and the correlation analysis results showed that P flux was significantly positively correlated with pH (p < 0.01), which further explained the relatively high P fluxes in winter and spring. However, the P release from sediments is not exactly the result of abiochemical processes mediated by physical conditions of water columns, but rather of complex interactions between biological and abiotic processes in water columns and sediments (Andersen, 1982; Nowlin et al., 2005). For example, high  $NO_3^- - N$  concentrations in the overlying water can raise the ORP by providing denitrifying bacteria with an alternate electron acceptor under anoxic conditions (Nowlin et al., 2005), thereby enhancing iron oxidation and phosphate adsorption. Notably, SRP flux had a significant (p < 0.05) negative correlation with  $NO_3^- - N$  in overlying water, implicating that high nitrate concentrations may inhibit P release from sediments to some extent, even under the poor oxidation conditions at the SWI in summer (Fig. 4).

It is well known that water temperature is an important factor

affecting N and P release from sediment (Jeppesen et al., 2005; Paytan et al., 2017). In this study, low-temperature water was discharged from the hypolimnion of the upstream PJKR in order to suppress the algal bloom. Except for winter, the surface water temperature was much lower in the shallow areas near inflows than in the deep areas downstream (Fig. S1). However, there was thermal stratification in the deep areas of DHTR during the warm seasons (Fig. S2), and minimal difference in WT between the hypolimnion of deep areas and the surface water in shallow water areas upstream. Therefore, in response to increasing water temperatures in the warm seasons, the stimulation of sediment N and P release was weaker in DHTR than in the shallow lakes. In summer, affected by heavy rains and upstream reservoir large discharge, part of water bodies developed vertically mixing, thus the DO stratification could be weakened (Fig. S2). Previous studies have indicated that compared with other environmental factors, DO is the dominant environmental factor affecting the P release from sediments (Nowlin et al., 2005; Ahlgren et al., 2011; Anderson et al., 2021; Sun et al., 2022). Even if the thermal stratification existed in summer, the hypolimnion was not in an anoxic condition; therefore, the dissolution and release potential of sediment SRP were relatively weak.

The spatial pattern of  $NH_4^+ - N$  fluxes resembled that of SRP fluxes. Both exhibited a gradual increasing trend from the shallow areas near inflows to the deep areas near the dam. There was a positive correlation between the  $NH_4^+ - N$  and SRP fluxes (Table S4), indicating that the release of N and P from sediments was synergistic. The contents of TN, inorganic N, labile-P, and organic matter in sediment were higher in deeper areas (Fig. 3). Organic matter decomposition is the main source of nutrients (Gibbons and Bridgeman, 2020). The higher degree of sediment pollution was, the higher mobile components of N and P, thus inducing higher rates of N and P release from sediment (Yang et al., 2020). Discharge from the upstream reservoir brings a mass of organic particles, and the increased delivery to sediment can lead to high burial and sedimentation rates (Friedl and Wuest, 2002). In addition, the reservoir hydrodynamics gradually weakened from upstream to downstream, forming a hydrostatic effect of dam. The hydrostatic effect in the deeper part promotes algae growth and burial of organic matter, which could lead to a decrease in the DO concentration in bottom water columns (Chen et al., 2018). In this study, the DO concentration at the SWI was apparently lower in deeper areas (Fig. 4). As a result, the P retention ability of sediment would have been diminished and  $NH_4^+ - N$  accumulation would have been promoted. It should be pointed out that in the shallow areas, due to the influence of the discharge of upstream reservoir, the surface water temperatures have been maintained at a low level in warm seasons (Fig. S1), and the microbial activities and organic matter decomposition will be inhibited by low temperature (Zhong et al., 2021), which will also contribute to lower N and P flux in the upstream areas.

## 4.4. Implications for eutrophication management of DHTR and other cascade reservoirs

Understanding the relative importance of internal and external nutrient sources is of great significance for eutrophication management. Based on the release rates of  $NH_4^+ - N$  and SRP across the SWI and the area of DHTR (30  $km^2$ ), the estimated annual  $NH_4^+ - N$  and SRP fluxes were 154.94 t and 54.31 t, respectively. The external nutrient sources of DHTR mainly included the water discharge from the upper PJKR, Sahe River inflows, and non-point source pollution from the surroundings of DHTR, for which N loadings were 4,997.64 t, 603.94 t, and 222.05 t, respectively, and P loadings were 86.79 t, 4.53 t, and 10.94 t, respectively in 2018 (Estimation methods are shown in the Supplementary Materials). The nutrient loadings of DHTR were mainly from external sources, which were primarily disturbed by the discharge water from the PJKR, especially for N. P was more strongly affected by internal loading release than N (Fig. 9). Therefore, attention should be paid to both

external and internal sources in the eutrophication management of DHTR. In the present study, the internal P loading accounted for 34.7% of the total P input. This contribution is higher than that in Lake Erie and four lakes in Eastern Nebraska (Paytan et al., 2017; Song et al., 2017), but lower than that in Lake Winnipeg (Nürnberg and LaZerte, 2016). This result is likely to have been attributed to the specific conditions of the water body and the experimental methods. It must be pointed out that the flux estimated in this study was based on the concentration gradient of pore water. Compared with the intact sediment core incubation method, the results of this study would have underestimated the internal loadings (Wen et al., 2019b), especially for N. Additionally, the N budget at the SWI is regulated by multiple N cycle processes (Zhong et al., 2021), but only  $NH_4^+ - N$  flux was estimated in this study. Thus, our future investigations should comprehensively evaluate the source–sink effect of sediments on N.

Based on the above results, we suggest that the water quality of PJKR should be improved to reduce the nutrient loadings in the upstream reservoir, sewage discharge in residential areas near the reservoir bank should be supervised, and ecological restoration can be implemented in the shallow water areas of Sahe River inflows. The N loading of DHTR mainly came from the discharge from the upstream reservoir; therefore, the N loading treatment of upstream reservoir is important and necessary in the cascade reservoirs. Recently, research into aerobic denitrifying bacteria in natural water bodies has received increasing attention, and a combination of aeration technology and aerobic denitrifiers can be used in situ to treat N loadings in reservoirs (Huang et al., 2022; Zhang et al., 2023). Compared with N loading, internal P loading is the main target of internal nutrient loading management. To effectively control P loading in DHTR, we further suggest reducing the internal P loading by chemical P-fixation (Lurling and Faassen, 2012; Yin et al., 2016), sediment dredging (Reddy et al., 2007; Yu et al., 2017), and hypolimnetic aeration (Dittrich et al., 2011). Our results show that the water quality of the cascade reservoir was greatly affected by that of the upstream reservoir. The low-temperature water discharged from the hypolimnion of the upstream reservoir can reduce the surface water temperature of the downstream reservoir, so as to inhibit harmful algal blooms to a certain extent. However, if the water source of the upstream reservoirs has high N and P loadings, it can provide nutrients for algal blooms in the downstream reservoirs. Hence, managers of cascade reservoirs need to consider the advantages and disadvantages of release from upstream reservoirs.

#### 5. Conclusion

This work investigated the magnitude and patterns of the internal N and P loadings in DHTR, an intermediate reservoir of three cascade reservoirs, and evaluated the relative importance of internal and external sources. Our results indicated that DHTR suffered severe sediment pollution, which was mainly attributed to the extensive cage culture disturbance in the reservoir prior to 2018. N and P fluxes presented considerable spatiotemporal variability, with the highest fluxes in autumn and winter, respectively, and the lowest fluxes in summer. This result was attributed to the large water level gradients and the water discharged from the hypolimnion of the upstream PJKR. We found that both N and P fluxes showed an increase from shallow water to deep water areas, which was attributed to the high nutrient loadings in the sediments and the lower DO concentration caused by thermal stratification and hydrodynamic conditions in deep water areas. The comparison of N and P loadings between internal and external sources demonstrated that the water discharged from the upstream reservoir was the main input source of N and P. Meanwhile, the internal P loading also contributed to water eutrophication to a great extent. Therefore, the eutrophication management of cascade reservoirs should consider the comprehensive effects of both internal and external nutrient loadings, especially the discharge water quality from upstream reservoirs.

#### Credit author statement

Min Zhang: Conceptualization, Data curation, Visualization, Formal analysis, Writing-Original draft, Writing-review & editing. Shuailong Wen: Investigation, Methodology, Data curation, Validation. Tao Wu: Investigation, Supervision, Methodology, Project administration, Shaoming Wang: Conceptualization, Data curation, Validation, Resources, Xin Li: Investigation, Methodology, Data curation, Validation, Wanqing Gong: Investigation, Methodology, Data curation, Validation, Hongwei Wang: Investigation, Methodology, Data curation, Validation, Cheng Liu: Investigation, Conceptualization, Methodology, Resources, Jicheng Zhong: Conceptualization, Methodology, Writing-review & editing, Resources, Funding acquisition, Project administration.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.



Fig. 9. Percentages of internal and external sources of N and P loadings in Daheiting Reservoir (DHTR).

#### Data availability

The authors do not have permission to share data.

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#### Appendix A. Supplementary data

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