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Seasonal variations and controlling factors of nitrogen fluxes at the sediment-water interface in a semi-enclosed inland sea

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Abstract. Nitrogen fluxes across the sediment-water interface and nitrogen removal from sediments are essential components of the nitrogen cycle and ecosystem in semi-enclosed inland seas. However, the difficulty in observational sampling hinders the acquisition of continuous data necessary to understand their seasonal variations and underlying mechanisms. In response to this issue, we have developed a one-dimensional vertical model of the nitrogen cycle within sediments and used it to reproduce the seasonal changes of observed nitrogen fluxes in a typical semi-enclosed inland sea and investigate their controlling factors through sensitivity experiments. Model results indicate that 40% of particulate organic nitrogen (PON) settling into sediments is returned to the bottom water as dissolved inorganic nitrogen (DIN), while 30% is removed via N-loss flux (dinitrogen gas and nitrous oxide). Although PON flux is controlled by PON concentration in the bottom water, DIN and N-loss fluxes show temperature-driven seasonal variations, suggesting a decoupling between nitrogen return and PON input. Additionally, seasonal variations in oxygen penetration depth (OPD), ranging from 1 to 3 mm, also affect nitrogen fluxes. In nitrate-depleted sediments of semi-enclosed seas, the denitrification rate is no longer significantly higher than the anammox rate in the nitrogen removal.

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Keywords: sediment-water interface, semi-enclosed inland sea, nitrogen fluxes, sediment model, nitrogen removal

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1 Introduction

Sediments play a crucial role in regulating the storage and removal of nitrogen within semi-enclosed inland seas, which significantly influences the balance and stability of the ecosystem (Huettel et al., 2014; Zhang et al., 2020; Kalvelage et al., 2013; Thamdrup, 2012). On one hand, the sediment can bury substantial amounts of nitrogen, potentially on par with river inputs (Liu et al., 2021; Han et al., 2021; Lønborg and Markager, 2021). On the other hand, because of remineralization, the sediment continuously releases dissolved inorganic nitrogen (DIN) into the overlying water (Devol, 2015; Boynton et al., 2017; Liu et al., 2012). In the semi-enclosed inland sea, vertical mixing facilitates the contribution of DIN from sediments as a vital nutrient source for primary production because of shallow water depth (Huettel et al., 2014; Yi et al., 2023; Leng et al., 2023; Liu and Yin, 2007; Berelson et al., 2013; Leynaert et al., 2011; Mei Liu et al., 2011). Notably, the sediment serves as the primary anoxic environment in these regions, where key nitrogen removal processes, such as denitrification and anammox, occur. These processes are essential for regulating nitrogen inventories and influencing nutrient structure in these areas. Researches indicate that continental shelves, which constitute merely 7.5% of the global seafloor, are responsible for 44% of fixed nitrogen losses (Devol, 2015; Mctigue et al., 2016; Huang et al., 2021; Jäntti and Hietanen, 2012; Khalil and Rasmussen, 2012). Furthermore, nitrous oxide, a byproduct of denitrification, is a significant greenhouse gas that contributes to global warming (Wilson et al., 2020; Yang et al., 2022).

To clarify the function of the sediment in ecological and environmental changes, it is necessary to conduct a quantitative analysis of nitrogen fluxes at the sediment-water interface in semi-enclosed inland seas, along with their seasonal variations. Direct observation of particulate organic nitrogen (PON) flux at this interface poses challenges due to the influence of resuspension processes. Consequently, numerous studies focused on DIN flux and its seasonal variations primarily through in situ observations, incubation experiments, or estimations based on Fick's law (De Vittor et al., 2012; Mu et al., 2017; Chen et al., 2023). For example, studies in the northern Adriatic Sea and the Baltic Sea have demonstrated the same conclusion that DIN flux exhibits seasonal variations, with higher levels in summer and lower levels in winter, likely influenced by water temperature (De Vittor et al., 2012; Niemistö et al., 2018). The fluxes of dinitrogen gas and nitrous oxide are mainly accessed through the measurement of denitrification and anammox rates within sediments (Zhang et al., 2018; Rich et al., 2020). A study conducted in the Pearl

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River Estuary reported that the denitrification rate in sediments also displayed seasonal variability, with peak values observed in summer and reduced values in winter (Teng and Lin, 2024). However, the challenges and cost associated with field sampling limit the feasibility of continuous monitoring of these fluxes. Moreover, the direction of DIN flux varies across different regions, with the underlying mechanisms remaining unclear (Mu et al., 2017; De Vittor et al., 2015; Dale et al., 2022; Bohlen et al., 2011; Ratmaya et al., 2022; Zhou et al., 2022). Therefore, identifying the magnitude and direction of nitrogen fluxes at the sediment-water interface in semi-enclosed inland seas, along with their seasonal patterns and underlying controlling mechanisms, is crucial for understanding the contribution of sediments to the aquatic ecosystem in this region.

Numerical modeling is a valuable tool for understanding the factors that govern nitrogen fluxes at the sediment-water interface. Typically, nitrogen fluxes at this interface derived from empirical formulas serve as essential boundary conditions for modeling the aquatic nitrogen cycle in semi-enclosed inland seas (Lønborg and Markager, 2021). However, this approach largely overlooks the changes occurring within the sediment and the corresponding flux responses to these alterations. An alternative approach is the implementation of a box model, which investigates nitrogen processes within the sediment. Nonetheless, the box model's treatment of all sediment as a single domain, coupled with its failure to account for the vertical distribution of chemical substances—particularly oxidants such as dissolved oxygen (DO)—limits its capacity to provide a comprehensive understanding of the nitrogen cycling processes within the sediment (Yang et al., 2022). A one-dimensional vertical sediment model addresses this limitation, albeit at the cost of increased complexity and computational demands. This model has demonstrated efficacy in accurately reproducing seasonal variations in nitrogen fluxes at the sediment-water interface (Radtke et al., 2019; Umlauf et al., 2023). Despite the clear advantages of this modeling approach over empirical formulas and box models, well-validated applications remain relatively scarce.

This study focuses on the unclear seasonal variations in the magnitude and direction of nitrogen fluxes at the sediment–water interface in semi-enclosed inland seas, alongside the insufficiently understood underlying mechanisms with controlling factors driving these changes. We developed a one-dimensional vertical sediment model to simulate nitrogen cycling and accurately quantify nitrogen fluxes at the interface. Supported by monthly continuous observations conducted in the sediment of Harima Nada in Japan, a typical semi-enclosed inland sea, we obtained a robust dataset for model validation. The model

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successfully reproduced the seasonal variations in the concentrations of three nitrogen species within the sediment and nitrogen fluxes at the interface. Additionally, numerical experiments further elucidated the influence of key environmental factors on these seasonal variations. This study aspires to establish a scientific foundation for comprehending the role of sediment nitrogen cycling in influencing the ecological environment of semi-enclosed marine systems. The findings can provide crucial support for offshore environmental remediation and the development of management policies.

2 Materials and Methods

2.1 Model descriptions

Based on the theory of early diagenesis (Berner, 1980), we have developed a biogeochemical model for sediment (Figure 1). This model encompasses two components: solid matter and pore water. The state variables consist of the concentrations of two types of PON (fast-decayed and slow-decayed ones) for the solid matter and the concentrations of NH_4^+ and NO_3^- in the pore water. Additionally, we set a fluff layer at the top of sediment, where the active benthos and material exchange processes with the above water column occur (Lee et al., 2002; Kuhrts et al., 2006; Laima et al., 2002).

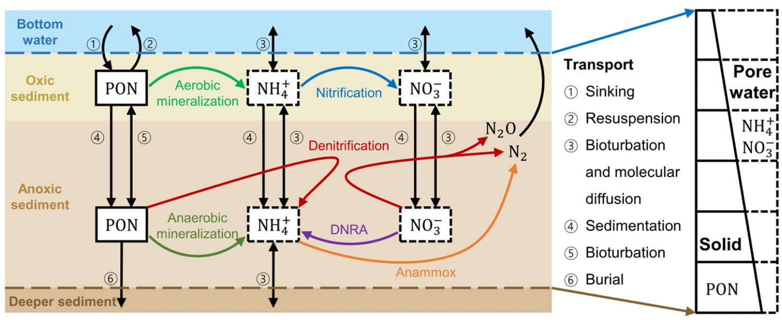

Figure 1. Physical and biogeochemical processes for nitrogen cycle in the sediment model. Three state variables in the model are PON, NH_4^+ and NO_3^-. PON is a solid substance (enclosed by solid lines), while NH_4^+ and NO_3^- are dissolved substances in the pore water (enclosed by dash lines). The physical processes and biogeochemical processes are denoted by black arrows and colored arrows, respectively. The numbers denote the transport processes included in the model.

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In the solid part, the fast-decayed and slow-decayed PON are treated with a large and small mineralization rate, respectively. Both of them undergo identical vertical transport processes, including downward sedimentation driven by gravity and vertical diffusion caused by bioturbation. Their mineralization occurring within the sediment provides a flux of NH_4^+ into the pore water.

In the pore water, the dissolved NH_4^+ and NO_3^- are transported downward and diffused vertically, as corresponding to both bioturbation and molecular diffusion. The biogeochemical processes occurring within the pore water include nitrification, denitrification, anammox and dissimilatory nitrate reduction to ammonium (DNRA).

At the sediment-water interface, the exchange of solid matter is controlled by the bottom shear stress. The solid matter in the water column sinks into the fluff layer when the bottom stress is below a critical value, while resuspension occurs when the bottom stress exceeds this threshold. The exchange fluxes of NH_4^+ and NO_3^- at the interface are determined by the product of the diffusion coefficient and their concentration gradient between the fluff layer and the overlying water.

In the deepest layer of sediment, only the slow-decayed PON is allowed to move downward and leave the model domain via burial. The exchange flux of NH_4^+ between the model domain and the underlying pore water is calculated by the production of the diffusion coefficient and the concentration gradient derived from observations, while the exchange flux of NO_3^- there is set to zero.

2.2 Equations and parameters

The equations for the concentrations of solid matter (C_s) and dissolved matter (C_d) are as follows:

$$\frac{\partial[(1-\emptyset)\cdot C_s]}{\partial t} = \frac{\partial[(1-\emptyset)\cdot D_B \cdot \frac{\partial C_s}{\partial z}]}{\partial z} - \frac{\partial[\omega\cdot(1-\emptyset)\cdot C_s]}{\partial z} + (1-\emptyset)\cdot\sum R_s \tag{1}$$

$$\frac{\partial[\emptyset\cdot C_d]}{\partial t} = \frac{\partial[\emptyset\cdot(D_B+D_M)\cdot\frac{\partial C_d}{\partial z}]}{\partial z} - \frac{\partial[\omega\cdot\emptyset\cdot C_d]}{\partial z} + \emptyset\cdot\sum R_d \tag{2}$$

Where z is vertical axis for sediment depth, t is time, $\emptyset$ is the sediment porosity, D_B and D_M are the bioturbation coefficient and molecular diffusion coefficient ($m^2\ s^{-1}$), ω is the sedimentation rate ($mm\ yr^{-1}$), R_s is the biochemical reaction rate of PON, and R_d is the biochemical reaction rate of dissolved matter, including NH_4^+ and NO_3^-. Since the sediment type in the study area is muddy, the advection of pore water in sandy sediments controlled by pressure is neglected.

The vertical profile of D_B is expressed as equation (3) (Radtke et al., 2019).

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$$D_B(z) = \begin{cases} D_{B_{max}} & , \ 0 < z < z_{max} \\ D_{B_{max}} \cdot \exp\left(-\frac{z - z_{max}}{z_d}\right), & z \geq z_{max} \end{cases} \tag{3}$$

Where z_{max} is the depth down to which the maximum bioturbation coefficient ($D_{B_{max}}$) is applied, z_d is the decaying length scale of $D_{B_{max}}$.

D_M of dissolved matter in the pore water is a function of porosity, and is expressed as follows (Boudreau et al., 1998):

$$D_M(z) = \frac{D_0}{1 - 2.02 \ln(\emptyset)} \tag{4}$$

where D_0 is the diffusion coefficient of the dissolved matter in the particle-free liquid ($m^2\ s^{-1}$).

The biochemical reactions are expressed in equations (5)-(7). Detailed descriptions of these reactions are presented in Table 1 (Soetaert et al., 1996; Capet et al., 2016; Akbarzadeh et al., 2018). These reaction equations primarily consider the limitations imposed by DO and temperature on each reaction. Due to the lack of parameterization schemes, it is difficult to represent the competition between different organisms for the same reaction. Since this is not the focus of this paper, we describe them collectively as a unified, generalized concept based on reaction rates. All the related coefficients and parameters are listed in Table 2. The sensitivity analysis of the model parameters is shown in Figure S1.

$$\sum R_{PON} = -R_{min}^{O_2} - R_{den} - R_{min}^{AM} - R_{DNRA} \tag{5}$$

$$\sum R_{NH_4^+} = R_{min}^{O_2} + R_{den} + R_{min}^{AM} + (1 + 0.5 \cdot r_{C:N}) R_{DNRA} - R_{nit} - R_{Ana} \tag{6}$$

$$\sum R_{NO_3^-} = R_{nit} - 0.5 \cdot r_{C:N} \cdot R_{DNRA} - 0.8 \cdot r_{C:N} \cdot R_{den} \tag{7}$$

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Table 1. Reaction formulas and rate expressions.

Process	Formula	Rate expression
Aerobic mineralization	$(CH_2O)_x(NH_3)_y(H_3PO_4)_z + xO_2 \rightarrow yNH_4^+ + xH_2O$	$R_{min}^{O_2} = r_{min} \cdot PON \cdot Q^{\frac{T-20}{10}} \cdot \frac{O_2}{O_2 + Ks_{O_2}} \cdot \frac{1}{\sum lim}$
Denitrification	$(CH_2O)_x(NH_3)_y(H_3PO_4)_z + 0.8xNO_3^- \rightarrow yNH_4^+ + 0.4x(1-\alpha)N_2 + 0.4x\alpha N_2O + 1.4xH_2O$	$R_{den} = r_{min} \cdot PON \cdot Q^{\frac{T-20}{10}} \cdot \frac{NO_3^-}{NO_3^- + Ks_{NO_3^-}} \cdot \left(1 - \frac{O_2}{O_2 + Kin_{O_2}^{Den}}\right) \cdot \frac{1}{\sum lim} \cdot \gamma$
Anaerobic mineralization	$(CH_2O)_x(NH_3)_y(H_3PO_4)_z$ + an oxidant $\rightarrow yNH_4^+ + xODU + xH_2O$	$R_{min}^{AM} = r_{min} \cdot PON \cdot Q^{\frac{T-20}{10}} \cdot \left(1 - \frac{O_2}{O_2 + Kin_{O_2}^{AM}}\right) \cdot \left(1 - \frac{NO_3^-}{NO_3^- + Kin_{NO_3^-}}\right) \cdot \frac{1}{\sum lim}$
Nitrification	$NH_4^+ + 2O_2 \rightarrow NO_3^- + H_2O$	$R_{nit} = r_{nit} \cdot NH_4^+ \cdot \frac{O_2}{O_2 + Ks_{Nit}}$
DNRA	$(CH_2O)_x(NH_3)_y(H_3PO_4)_z + 0.5xNO_3^- \rightarrow (0.5x + y)NH_4^+$	$R_{DNRA} = r_{min} \cdot PON \cdot Q^{\frac{T-20}{10}} \cdot \frac{NO_3^-}{NO_3^- + Ks_{NO_3^-}} \cdot \left(1 - \frac{O_2}{O_2 + Kin_{O_2}^{Den}}\right) \cdot \frac{1}{\sum lim} \cdot (1-\gamma)$
Anammox	$NH_4^+ + NO_2^- \rightarrow N_2 + 2H_2O$	$R_{Ana} = r_{Ana} \cdot \frac{NH_4^+}{NH_4^+ + Ks_{NH_4^+}} \cdot \frac{NO_2^-}{NO_2^- + Ks_{NO_2^-}} \cdot \frac{Kin_{O_2}^{Ana}}{O_2 + Kin_{O_2}^{Ana}}$

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$$\sum \lim = \frac{O_2}{O_2 + K_{O_2}} + \frac{NO_3^-}{NO_3^- + Ks_{NO_3^-}} \cdot \left(1 - \frac{O_2}{O_2 + Kin_{O_2}^{Den}}\right) + \left(1 - \frac{O_2}{O_2 + Kin_{O_2}^{AM}}\right) \cdot \left(1 - \frac{NO_3^-}{NO_3^- + Kin_{NO_3^-}}\right).$$

$x : y : z =$ C: N: P.

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162 **Table 2.** Model parameters.

Parameter	Value	Unit	Description	References
ω	2.6	mm yr^{-1}	Sedimentation rate	(Ichimi et al., 2005)
$D_{B_{max}}$	1.15×10^{-11}	m^2 s^{-1}	Maximum bioturbation coefficient	(Soetaert et al., 1996)
z_{max}	0.01	m	Depth down to which the Maximum bioturbation coefficient is applied	(Soetaert et al., 1996)
z_d	0.5	m	Decaying length scale of the maximum bioturbation coefficient	(Soetaert et al., 1996)
$D_0^{NH_4^+}$	0.847	cm^2 d^{-1}	The molecular diffusion coefficient of ammonium in a particle-free solution at 0 °C	(Soetaert et al., 1996)
$D_0^{NO_3^-}$	0.845	cm^2 d^{-1}	The molecular diffusion coefficient of nitrate in a particle-free solution at 0 °C	(Soetaert et al., 1996)
r_{minf}	2.8×10^{-9}	s^{-1}	Mineralization rate of fast decay organic nitrogen	
r_{mins}	1.05×10^{-9}	s^{-1}	Mineralization rate of slow decay organic nitrogen	
Q	2		Factor for temperature effect on mineralization	(Capet et al., 2016)
Ks_{O_2}	3	µmol L^{-1}	Half saturation constant of oxygen in aerobic mineralization	(Soetaert et al., 1996)

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$Ks_{NO_3^-}$	30	µmol L^{-1}	Half saturation constant of nitrate in denitrification	(Soetaert et al., 1996)
$Kin_{O_2}^{Den}$	10	µmol L^{-1}	Half saturation constant of oxygen in denitrification	(Soetaert et al., 1996)
γ	95	%	Fraction of total nitrate Reduction occurring via denitrification	(Akbarzadeh et al., 2018)
$Kin_{O_2}^{AM}$	5	µmol L^{-1}	Half saturation constant of oxygen in anaerobic mineralization	(Soetaert et al., 1996)
$Kin_{NO_3^-}$	5	µmol L^{-1}	Half saturation constant of nitrate in anaerobic mineralization	(Soetaert et al., 1996)
r_{nit}	20	d^{-1}	Maximum nitrification rate	(Soetaert et al., 1996)
Ks_{nit}	1	µmol L^{-1}	Half saturation constant of oxygen in nitrification	(Soetaert et al., 1996)
r_{Ana}	2.3×10^{-4}	mmol m^{-3} s^{-1}	Maximum anammox rate	(Akbarzadeh et al., 2018)
$Ks_{NH_4^+}$	5	µmol L^{-1}	Half saturation constant of ammonium in anammox	(Akbarzadeh et al., 2018)
$Ks_{NO_2^-}$	5	µmol L^{-1}	Half saturation constant of nitrite in anammox	(Akbarzadeh et al., 2018)
$Kin_{O_2}^{Ana}$	8	µmol L^{-1}	Half saturation constant of oxygen in anammox	(Akbarzadeh et al., 2018)

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$r_{C:N}$	10.2		Ratio of carbon to nitrogen	
α	1	%	Nitrous oxide leakage during denitrification	(Akbarzadeh et al., 2018)
w	2	m s^{-1}	Sinking rate	(Ding et al., 2020)
τ_c	0.06	N m^{-2}	Critical bottom stress	(Ding et al., 2020)
E_{PON}	1×10^{-8}	mmol m^{-2} s^{-1}	Resuspension coefficient	(Ding et al., 2020)
D_{sur}	7×10^{-7}	m^2 s^{-1}	Surface diffusion coefficient	(Radtke et al., 2019)
Δz	3	m	Distance of the diffusion layer at sediment-water interface	(Nakakuni et al., 2024)

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Due to the large biomass of benthos and high DO concentrations in the fluff layer, the mineralization rate in this layer is set to be 50 times faster than that in the sediment below it (Lee et al., 2002; Kuhrts et al., 2006; Laima et al., 2002).

At the sediment-water interface, the sinking and resuspension of PON depend on the bottom stress. The exchange flux (F_s) is expressed as (Wang, 2002):

$$F_s = \begin{cases} w \cdot C_s^w \cdot (\frac{\tau}{\tau_c} - 1), & \tau < \tau_c \\ E_s \cdot (\frac{\tau}{\tau_c} - 1), & \tau \geq \tau_c \end{cases} \quad (8)$$

where w is the sinking velocity of particles in the bottom water ($m\ s^{-1}$), C_s^w is concentration of PON in the bottom water ($mmol\ m^{-3}$), τ is the bottom stress ($N\ m^{-2}$), τ_c is the critical bottom stress ($N\ m^{-2}$), E_s is the resuspension rate ($mmol\ m^{-2}\ s^{-1}$).

The exchanges of dissolved matters (NH_4^+ and NO_3^-) are mainly through bioturbation and molecular diffusion, and the flux (F_d) is expressed as (Boudreau et al., 1998):

$$F_d = -D_{sur} \cdot \emptyset \cdot \frac{C_d^w - C_d^{pw}}{\Delta z} \quad (9)$$

where D_{sur} is the surface diffusion coefficient ($m^2\ s^{-1}$). C_d^w and C_d^{pw} are the concentrations of dissolved matter in the bottom water and pore water of fluff layer, respectively ($mmol\ m^{-3}$). Δz is the distance of the diffusion layer at sediment-water interface (m).

At the bottom of the model domain, the slow-decayed PON is allowed to move out as the burial process with a flux (B_s) as (Radtke et al., 2019):

$$B_s = \omega \cdot (1 - \emptyset) \cdot C_s^b \quad (10)$$

where superscript of b represents the deepest layer of the model domain.

The bottom flux of NH_4^+ (B_d) is expressed as:

$$B_d = -(D_B^b + D_M^b) \cdot \emptyset \cdot \frac{C_d^b - C_d^{bc}}{\Delta z^b} \quad (11)$$

where C_d^{bc} is NH_4^+ concentration in the pore water under the deepest layer of the model domain ($mmol\ m^{-3}$).

2.3 Study area

Harima Nada is situated within Seto Inland Sea, the largest semi-enclosed sea in Japan (Figure 2). It has an average water depth of 26 meters and is connected to Osaka Bay and Hiuchi Nada on the eastern and western sides, respectively. To the south, it connects to the Pacific Ocean through the Naruto Strait and

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Kii Channel (Zhu et al., 2019; Chang et al., 2009). Over the past few decades, the environment of the Seto Inland Sea has undergone significant changes (Ishii et al., 2014; Yamamoto et al., 2021). With the continuous observations of sediments in Harima Nada by our collaborators, we have realized that the DIN input from sediments in this area is comparable to that from rivers, exhibiting notable seasonal variations (Leng et al., 2023; Nakakuni et al., 2024). However, the driving factors and mechanisms behind these seasonal changes remain unclear. Therefore, Harima Nada serves as an excellent study area, and these observational data can provide a sufficient validation to ensure the reliability of model results (Nakakuni et al., 2024). This allows us to analyze the seasonal variations in sediment nitrogen cycling and interfacial nitrogen fluxes, as well as their controlling mechanisms. Additionally, this study lays the groundwork for future exploration of how sediment nitrogen cycling responds to long-term changes in the aquatic environment.

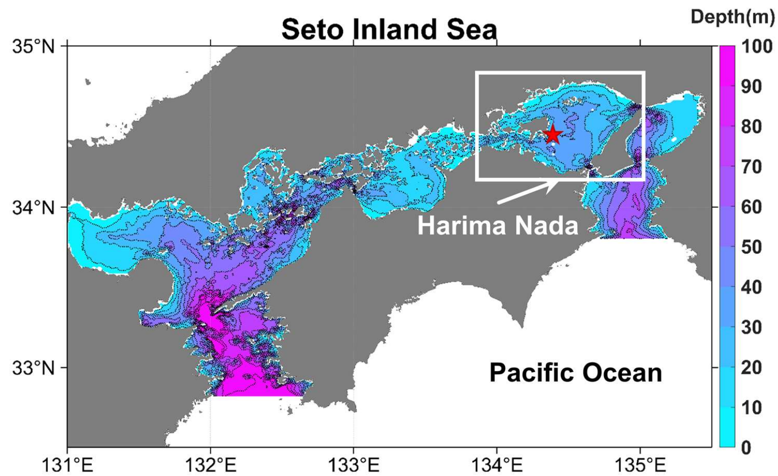

Figure 2. Study area of the Harima Nada, Japan. The red star represents the site where the observational data used in this study were collected.

2.4 Model configurations

Since this study focuses on seasonal changes at the sediment–water interface, the sediment depth in the model was set to 10 cm and uniformly divided into 100 layers.

The initial concentrations of the four state variables were obtained from the core observations in Harima Nada in July 2020 (Nakakuni et al., 2024). Moreover, the observed profile of DO concentration (Sayama

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et al., 2002), nitrite (NO_2^-) concentration, and porosity were also included and always constant in the model as input condition (Figure S2c-e and Text S2).

Monthly observations of bottom water temperature and concentrations of PON, NH_4^+, and NO_3^- from February 2020 to January 2021 were used to prescribe the upper boundary conditions, while NH_4^+ concentration beneath the model domain from April 2020 to May 2021 served as the bottom boundary conditions (Nakakuni et al., 2024). The bottom stress was derived from a hydrodynamic model for the Seto Inland Sea (Zhu et al., 2019) (Figure 3). Forced by these boundary conditions, the sediment model was calculated for a long time (more than 500 years) to reach a steady-state condition, wherein the input and output fluxes of materials become balanced.

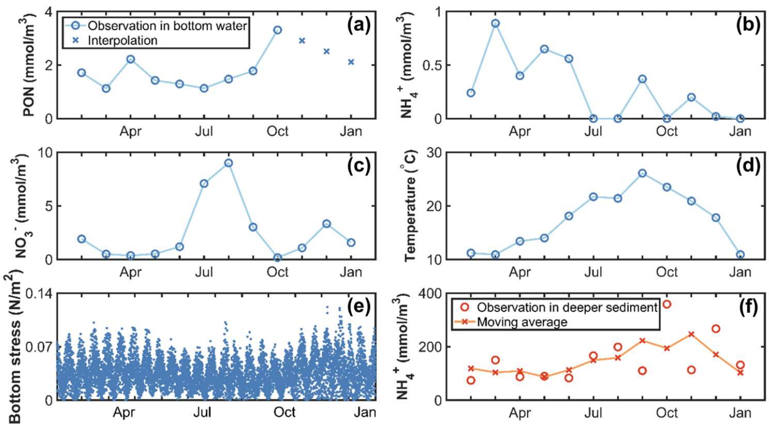

Figure 3. Boundary conditions of the model: (a) PON concentration, (b) NH_4^+ concentration, (c) NO_3^- concentration in the bottom water, (d) bottom water temperature, (e) bottom stress and (f) NH_4^+ concentration in the layer below model domain (10-12 cm). The values of PON concentration in the bottom water from November to January are obtained by linear interpolation. A three-point moving average is applied to NH_4^+ concentrations in (f) to smooth out intra-seasonal fluctuations.

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3 Results

3.1 Seasonal variations of the particle and dissolved nitrogen concentrations in the sediment

The monthly concentrations of PON, NH_4^+ and NO_3^- at different layers from 0 to 10 cm in the model domain are shown in Figure 4, alongside the corresponding observed data. The comparison between the model results and the observations, especially for the data in July (Figure 4d-e), suggests a reliable performance of the model.

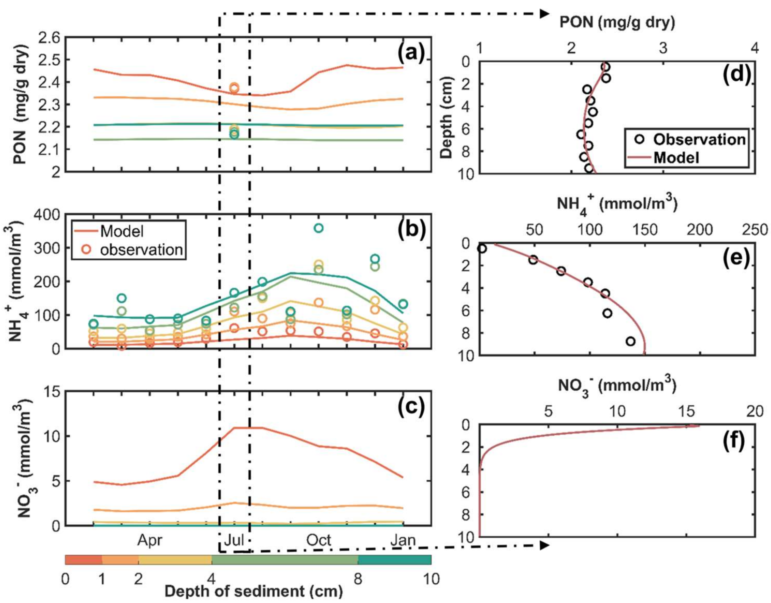

Figure 4. The seasonal variations of concentrations of (a) PON, (b) NH_4^+ and (c) NO_3^- in different layers in the sediment, the vertical profiles of (d) PON, (e) NH_4^+ and (f) NO_3^- in July. Colors in (a)-(c) represent different depth. In all the panels, lines are model results and circles are the observation data.

The concentration of PON in the sediment was consistently greater in the surface layer compared to the underlying layers (Figure 4a). Its seasonal variation was apparent in the upper 3 cm of sediment and almost disappears in the layers below 3 cm. In the 0-1 cm layer, PON concentration decreased from a value of 2.46 mg/g in February to a minimum value of 2.34 mg/g in August, followed by an increase to a peak concentration of 2.47 mg/g in November. In the deep layer of the sediment (8-10 cm), PON concentration remained around 2.20 mg/g throughout the year.

The NH_4^+concentration had a minimum value at the surface and a maximum value at the deepest layer (Figure 4b), which was contrary to the vertical distribution of PON concentrations. The NH_4^+

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concentrations presented significant seasonal variations across entire sediment layers, with the range of variation increasing with depth. For example, the NH_4^+ concentration in the 0-1 cm layer presented the lowest value of 11.02 mmol m^{-3} in February and the highest value of 38.71 mmol m^{-3} in September; however, the NH_4^+ concentration in the deep layer of the sediment (8-10 cm) presented the lowest value of 90.97 mmol m^{-3} in April and the highest value of 224.34 mmol m^{-3} in September. Except for the episodic high values observed in the deep sediment layer during autumn, the model successfully reproduced the seasonal variation of NH_4^+ concentration across all sediment layers, which was characterized by lower levels in winter and spring and elevated levels in summer and autumn.

NO_3^- concentration reached its peak at the sediment surface and exhibited a rapid decline to zero at the depth of around 3 cm (Figure 4c and f). Its seasonal variation occurred within the upper 1 cm layer, showing a high value in summer. The maximum concentration in the 0-1 cm layer was 10.91 mmol m^{-3} in July, which was double the minimum concentration of 4.56 mmol m^{-3} in March.

3.2 The flux of PON, DIN and N-loss (N_2 and N_2O) between bottom water and sediment

PON sank from the bottom water to the sediment throughout the year (Figure 5a). PON flux was high in fall with the maximum value of 2.90 mmol m^{-2} d^{-1} in October and low in spring with the minimum value of 1.07 mmol m^{-2} d^{-1} in May.

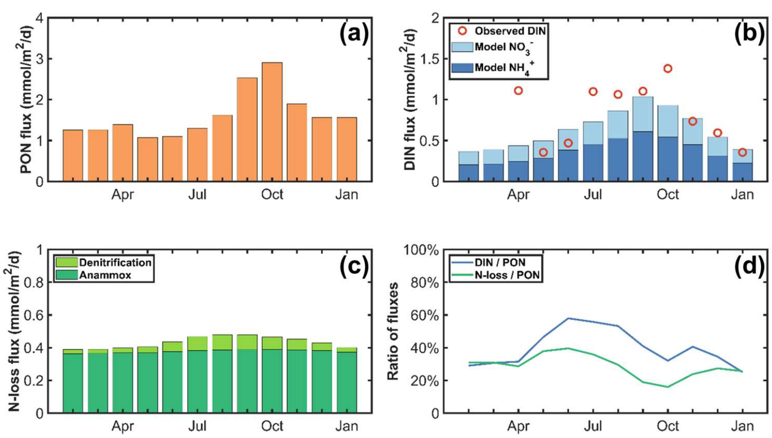

Figure 5. The fluxes of (a) PON, (b) DIN, (c) N-loss (N_2 and N_2O) at the sediment-water interface in the model, and (d) the ratios of DIN and N-loss fluxes to PON flux. The red circles in (b) are the values given by observations.

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The fluxes of NH_4^+ and NO_3^- was summed up to account the flux of DIN between bottom water and sediment. DIN in the Harima Nada was released from the sediment into the bottom water throughout the year, exhibiting the highest flux of 1.04 mmol m^{-2} d^{-1} in September and the lowest flux of 0.37 mmol m^{-2} d^{-1} in February (Figure 5b). The DIN flux and its seasonal variation given by the model exhibit a general agreement with observations (Nakakuni et al., 2024), although the model does not capture the observed high flux in April. The NH_4^+ and NO_3^- fluxes demonstrated comparable seasonal trends, reaching their respective maxima in September (0.61 and 0.43 mmol m^{-2} d^{-1}) and minima in February (0.20 and 0.16 mmol m^{-2} d^{-1}).

N-loss is produced through denitrification and anammox inside the sediment, which releases N_2 and N_2O to the atmosphere. N-loss flux across the sediment-water interface reached its peak of 0.48 mmol m^{-2} d^{-1} in August and the lowest value of 0.39 mmol m^{-2} d^{-1} in February (Figure 5c). The anammox was the main process for producing N_2 and contributed a value of around 0.37 mmol m^{-2} d^{-1} to N_2 flux, with minimal seasonal variation. Conversely, denitrification contributed to a seasonal variation of N-loss flux, reaching a maximum of 0.09 mmol m^{-2} d^{-1} in August and a minimum of 0.03 mmol m^{-2} d^{-1} in March. Although N_2 was the main product of denitrification, the flux of N_2O, a potent greenhouse gas, was not negligible. It varied from 0.3 to 0.9 μmol m^{-2} d^{-1}, with a mean value of 0.55 μmol m^{-2} d^{-1}.

The ratio of DIN flux to PON flux ($R_{DIN/PON}$) reflects the fraction of nitrogen that sinks into the sediment and later releases back to the seawater (Figure 5d). $R_{DIN/PON}$ increased from 25% in January to a peak of 58% in June, with the annual average of 40%. The ratio of N-loss flux to PON flux ($R_{N\text{-}loss/PON}$) represented the proportion of nitrogen removed from the sediment. The highest value of $R_{N\text{-}loss/PON}$ was 40% in June while the lowest value was 16% in October. The sum of $R_{DIN/PON}$ and $R_{N\text{-}loss/PON}$ ranged from 48% in October to 98% in June, with an annual average of 69%. It suggested that around 30% of the PON flux sinking from seawater was retained in the sediment on average, with this retention rate potentially reaching nearly 50% during certain months.

3.3 The budgets of PON, NH_4^+ and NO_3^- in the sediment

The annual budgets for the three forms of nitrogen in the sediment of Harima Nada are estimated from the model results (Figure 6). The sinking of PON from bottom water was the source of PON in the sediment, quantified at 8.30 g N m^{-2} yr^{-1}. Since the primary production in the Harima Nada in 2020 was

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estimated to be 41 g N m^{-2} yr^{-1} (Tada, 2021), it could be interpreted that about 20% of the particles produced by primary production sank into the sediment. Within the deeper layers of the sediment, the burial rate of PON was 2.60 g N m^{-2} yr^{-1}, representing 31% of the PON flux into the sediment and 6% of the primary production. The rest 5.69 g N m^{-2} yr^{-1} of PON flux into the sediment was mineralized to NH_4^+, which became the main source of NH_4^+ in the sediment.

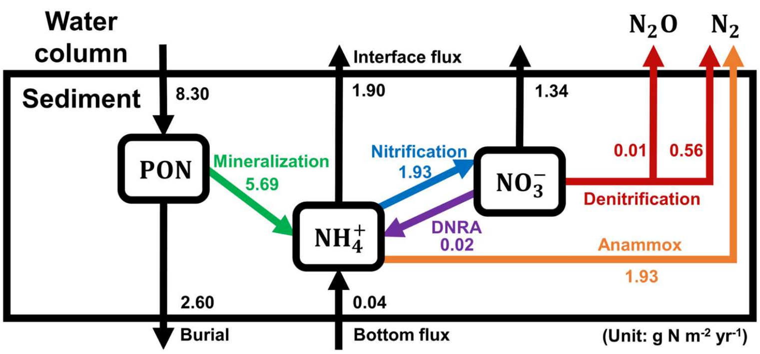

Figure 6. Nitrogen budgets in the sediment model of Harima Nada.

In addition to mineralization, DNRA also produced NH_4^+ in our model, but its value of 0.02 g N m^{-2} yr^{-1} was two orders of magnitude lower than the amount of mineralization (5.69 g N m^{-2} yr^{-1}). And 0.04 g N m^{-2} yr^{-1} of NH_4^+ was supplied from the deeper sediment located beneath the model domain. Furthermore, NH_4^+ has three sinks: the nitrification of NH_4^+ to NO_3^- had a rate of 1.93 g N m^{-2} yr^{-1}, the anammox of NH_4^+ to N_2 had a rate of 1.93 g N m^{-2} yr^{-1}, and the upward NH_4^+ flux through the sediment-water interface was quantified at 1.90 g N m^{-2} yr^{-1}.

For the NO_3^- produced by the nitrification, it mainly had an upward flux of 1.34 g N m^{-2} yr^{-1} at the sediment-water interface and the denitrification of 0.57 g N m^{-2} yr^{-1}. The combined effects of denitrification of NO_3^- and anammox of NH_4^+ produced a N-loss flux to the atmosphere amounting to 2.50 g N m^{-2} yr^{-1} (2.49 for N_2 and 0.01 for N_2O). Since this value represented over 30% of PON sinking flux to the sediment, it became an important process for removing nitrogen in the semi-enclosed inland sea.

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4 Discussion

4.1 Factors affecting the seasonal variations in the sediment-water fluxes of nitrogen

The model results show that the fluxes of PON, DIN and N-loss between seawater and sediment exhibit apparent seasonal variations. To clarify the factors controlling these seasonal variations, we carried out five sensitivity experiments, in which we removed the seasonal variation of one of the following five variables by substituting its monthly means with its annual mean: PON concentration (PON^{bw}), NH_4^+ concentration (${NH_4^+}^{bw}$), NO_3^- concentration (${NO_3^-}^{bw}$), bottom water temperature, and bottom stress. Figure 7 shows the calculation results of the five sensitivity experiments alongside the results described in Section 3 (referred to "Control" in Figure 7).

The concentration of PON^{bw} and the bottom water temperature significantly influence the seasonal variation of PON inventory in the sediment (Figure 7a). Sinking of PON^{bw} serves as the source of PON in the sediment, while variations in bottom water temperature can change the mineralization rate in the sediment (Figure S3a and Text S3). Although these seasonal variations can directly impact the PON inventory, the extent of seasonal variation in PON inventory is relatively minor, with its value being more than two orders of magnitude smaller than the overall PON inventory.

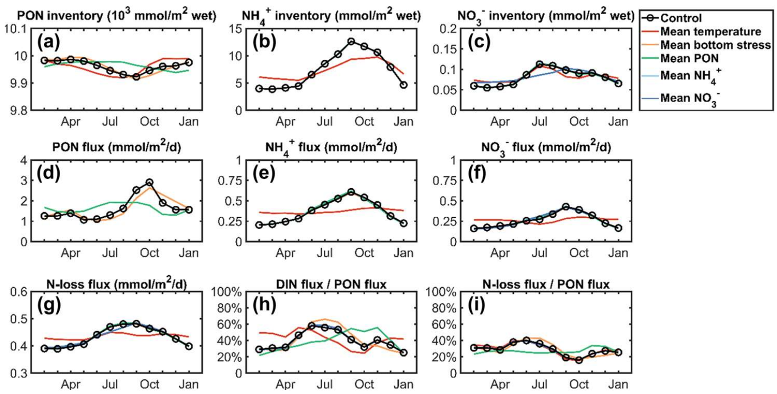

Figure 7. Comparisons of the model results among six calculations (five sensitivity experiments and one control run). The sensitivity experiments include the cases removing the seasonal variations of PON concentration (green line, "Mean PON"), NH_4^+ concentration (light blue, "Mean NH_4^+"), NO_3^- concentration (dark blue, "Mean NO_3^-"), and water temperature (red line, "Mean temperature") in the overlying water, as well as of the bottom stress (orange line, "Mean bottom stress"). (a) PON inventory; (b) NH_4^+ inventory; (c) NO_3^- inventory; (d) PON flux; (e) NH_4^+ flux; (f) NO_3^- flux; (g) N-loss flux; (h) ratio of DIN flux to PON flux; (i) ratio of N-loss flux to PON flux.

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Among the 5 factors, only the bottom water temperature is responsible for the seasonal variations of NH_4^+ inventory (Figure 7b) as it affects the mineralization rate that produces NH_4^+ in the sediment (Figure S3 and Text S3). The seasonal variation of NO_3^- inventory is mainly controlled by NO_3^{-bw} instead of bottom water temperature (Figure 7c). When the concentration of NO_3^{-bw} is fixed at its annual mean, a decrease in concentration enhances an increase in NO_3^- flux from the sediment to the overlying water (Figure 7f), leading to a corresponding decline in NO_3^- inventory (Figure S4 and Text S4).

The seasonal variation in concentration of PON^{bw} is the main factor deciding the seasonal variation of PON flux across the interface between the bottom water and sediment (Figure 7d). High concentration of PON^{bw} leads to an increased sinking flux of PON from seawater to sediment. With the annual average value of PON^{bw} concentration as the input condition, the peak PON flux from the seawater to sediment in October disappears (Figure 3a). In such a case, an inverse seasonal relationship can be discerned between PON flux (the green line in Figure 7d) and bottom stress (Figure 3e). The rise in bottom stress during the spring and winter induces the resuspension of PON at the sediment-water interface, thereby diminishing the downward PON flux.

The bottom water temperature controls not only the seasonal variations of NH_4^+ inventory in the sediment (Figure 7b) but also that of NH_4^+ flux from the sediment to the bottom water (Figure 7e). This is because NH_4^+ concentration in the sediment is much higher than the concentration of NH_4^{+bw}, and its variation directly determines the change of NH_4^+ flux (Figure 3b and Figure 4b). Since NH_4^+ is the sole source of NO_3^- in the sediment by nitrification (Figure 6 and Figure S3b), NO_3^- flux is consequently controlled by the bottom water temperature (Figure 7f). As a combination of NH_4^+ flux and NO_3^- flux, the seasonal variation of DIN flux is also controlled by the bottom water temperature, which is consistent with the conclusion suggested from field studies (Nakakuni et al., 2024).

The differences in the factors controlling the seasonal variation of PON and DIN fluxes indicate that a large PON flux into the sediment does not necessarily lead to a corresponding increase in DIN flux from the sediment to the bottom water. This suggests that estimating the DIN flux by using reflective boundary conditions based only on the concentration of PON^{bw}, or relying on empirical formulations that neglect bottom water temperature, may lead to considerable inaccuracies in the representing of the seasonal variation in DIN flux.

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N-loss occurs through denitrification and anammox, and is consequently closely associated with the inventories of NH_4^+ and NO_3^- in the sediment. Our calculation shows that the seasonal variation in N-loss flux mainly depends on the bottom water temperature and ${NO_3^-}^{bw}$ (Figure 7g). Since anammox is the dominant process for producing N_2 in the sediment, the bottom temperature, rather than ${NO_3^-}^{bw}$, has a greater impact on the seasonal variation of N-loss flux.

$R_{DIN/PON}$ and $R_{N\text{-}loss/PON}$ are two important parameters for comprehending the nitrogen cycle in the semi-enclosed inland sea. The seasonal variation of $R_{DIN/PON}$ is likely controlled by PON^{bw} and bottom temperature (Figure 7h). The annual mean of bottom temperature appears to advance the peak of $R_{DIN/PON}$, whereas the annual mean of PON^{bw} delays this peak. Conversely, $R_{N\text{-}loss/PON}$ is only affected by PON^{bw} (Figure 7i). The absence of an effect from bottom temperature effect on $R_{N\text{-}loss/PON}$ is caused by the relatively smaller range of N-loss flux compared to PON flux.

4.2 Influences of the DO profiles on nitrogen cycle in the sediment

The availability of DO has an important impact on the nitrogen cycle in the pore water. The DO concentration has a maximum value at the top of the pore water and diminishes with increasing depths (Devol, 2015). Consequently, the vertical profile of DO concentration in the sediment is characterized by a surface maximum concentration (DO_{max}) and the oxygen penetration depth (OPD). To assess the sensitivity of our model results to the specified DO concentration in the sediment, we conducted two groups of numerical experiments to clarify the effects of DO_{max} (GROUP I) and OPD (GROUP II) on the nitrogen cycle in the sediment.

According to the observed data obtained from the Ministry of the Environment of Japan (https://water-pub.env.go.jp/water-pub/mizu-site/mizu/kouiki/dataMap.asp), the average concentration of DO in the bottom water in the Seto Inland Sea ranged from 429 to 714 mmol m^{-3} in the past 40 years, which is far beyond the DO concentration in the fluff layer as the input data (84 mmol m^{-3}). Therefore, in GROUP I of the numerical experiments, DO_{max} was varied within the range of 60-140 mmol m^{-3}, while OPD was maintained at a constant value of 2 mm. The model results showed that there were no significant changes in the nitrogen inventories of the three types of nitrogen or their fluxes at the sediment-water interface (figures not given). The reason is that DO is not a limiting factor for nitrogen cycle within the aerobic zone of the upper 3 mm of sediment when DO_{max} is maintained between 60-140 mmol m^{-3}.

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It has been reported that the OPD in inland sediment can reach around 5 mm (Dale et al., 2011). In the five simulations of GROUP II, OPD was changed from 1 to 5 mm in 1 mm increments, while DO_{max} was maintained at a constant level of 80 mmol m^{-3}. The vertical profiles of DO concentration were derived through exponential fitting. The model results indicated that OPD has clear influences on NO_3^- inventory, NH_4^+ flux, NO_3^- flux and N-loss flux (Figures 8c, e, f, g). An increase of OPD facilitates the occurrence of nitrification in an expanded aerobic zone, leading to a greater conversion of NH_4^+ to NO_3^-. Since NH_4^+ mainly remains in the deeper sediment layers (Figure 4b), nitrification occurring in the surface sediment has a minimal impact on NH_4^+ inventory (Figure 8b). However, it can significantly reduce NH_4^+ flux at the sediment-water interface (Figure 8e). NO_3^- inventory in the sediment increases by 218% as OPD rises from 1 to 5 mm (Figure 8c). The increase in NO_3^- inventory subsequently enhances both NO_3^- flux and denitrification, resulting in a higher N-loss flux (Figures 8f and g). However, there is minimal difference in the NH_4^+, NO_3^-, and N-loss fluxes among the cases with OPD values of 3, 4, and 5 mm, indicating that beyond a certain critical threshold, the influence of OPD on these fluxes becomes negligible. Therefore, if the OPD exhibits seasonal variation between 1 and 3 mm, it will have a significant impact on the seasonal variation of nitrogen fluxes; if the OPD remains consistently deeper than 3 mm, its impact is less pronounced. Moreover, since an increase in OPD has opposing effects on NH_4^+ and NO_3^- fluxes, its overall influence on DIN flux is minimal.

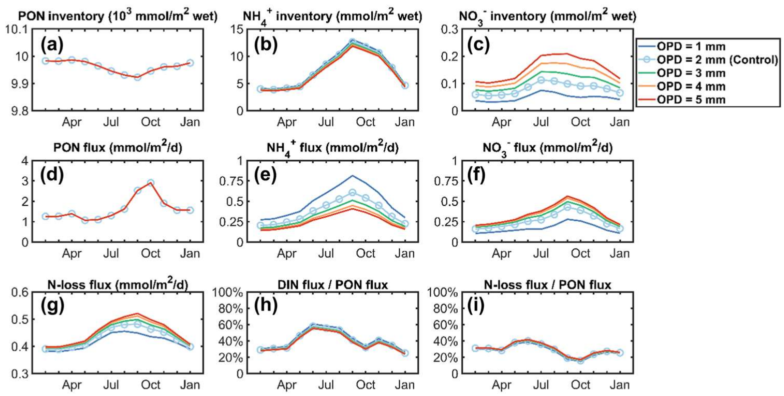

Figure 8. The effect of OPD depth on the nitrogen cycle in sediment: (a) PON inventory; (b) NH_4^+ inventory; (c) NO_3^- inventory; (d) PON flux; (e) NH_4^+ flux; (f) NO_3^- flux; (g) N-loss flux; (h) ratio of DIN flux to PON flux; (i) ratio of N-loss to PON flux.

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4.3 Comparisons with other coastal seas

Numerous studies have evaluated the nitrogen fluxes at the sediment-water interface, yielding varying conclusions (Zhou et al., 2017; Sun et al., 2021; Canion et al., 2014; Lin et al., 2017; De Vittor et al., 2012; Mu et al., 2017; Rich et al., 2020; Dale et al., 2022; Bohlen et al., 2011; Ratmaya et al., 2022; Zhou et al., 2022). In this subsection, we compare the fluxes of NH_4^+, NO_3^- and N-loss calculated by our sediment model with the values reported in these studies for different coastal seas (Table 3).

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Table 3. Benthic fluxes (mmol m^{-2} d^{-1}) reported in some regions; positive values represent the release from sediment to water.

Flux	Value		Location	References
NH_4^+	1.53	to 1.62	Northern Adriatic Sea	(De Vittor et al., 2012)
	0.16	to 1.39	Mauritanian upwelling region	(Dale et al., 2022)
	0.12	to 5.3	Vilaine Bay	(Ratmaya et al., 2022)
	-0.1077	to 0.219	Areas off eastern Taiwan	(Zhou et al., 2017)
	-0.71	to 0.14	Bohai Bay	(Mu et al., 2017)
	-0.1	to 0.066	Yellow Sea and East China Sea	(Zhou et al., 2022)
	0	to 4	Peruvian upwelling region	(Bohlen et al., 2011)
	0.0867	to 0.4722	Taranto Gulf	(De Vittor et al., 2015)
	0.22	to 0.63	Harima Nada	This study
NO_3^-	-0.46	to -0.26	Northern Adriatic Sea	(De Vittor et al., 2012)
	-3	to -0.1	Mauritanian upwelling region	(Dale et al., 2022)
	-0.62	to 0.46	Vilaine Bay	(Ratmaya et al., 2022)
	-0.241	to 0.00005	Areas off eastern Taiwan	(Zhou et al., 2017)
	1.43	to 2.26	Bohai Bay	(Mu et al., 2017)
	-3.43	to 0.082	Yellow Sea and East China Sea	(Zhou et al., 2022)
	-3.8	to -0.3	Peruvian upwelling region	(Bohlen et al., 2011)
	4.8	to 8.8	Taranto Gulf	(De Vittor et al., 2015)
	0.17	to 0.43	Harima Nada	This study
N_2	2.7	to 544.3	Chinese marginal seas	(Sun et al., 2021)

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28.70	to 420.49	East China Sea	(Lin et al., 2017)
0.2	to 2.8	Arctic shelf	(Canion et al., 2014)
1322.4	to 2683.2	Peru margin	(Rich et al., 2020)
0.40	to 0.48	Harima Nada	This study

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NH_4^+ is released from the sediment to the bottom water in our study area. It is commonly observed and has been reported in other coastal regions, including Northern Adriatic Sea (De Vittor et al., 2012), Vilaine Bay (Ratmaya et al., 2022) and Eastern Taiwan (Zhou et al., 2017). The difference between our study area and these coastal waters is that the sediment in our region also serves as a source of NO_3^- for the overlying seawater. The NO_3^- concentrations observed in the bottom water of Harima Nada were not always higher than that in the pore water. Consequently, NO_3^- produced by nitrification in the sediment can be released into the water column. In contrast, in areas such as the offshore of eastern Taiwan, the sediment continuously absorbs NO_3^- from the overlying water because of the lower concentration of NO_3^- in the pore water compared to the overlying water (Zhou et al., 2017).

The N-loss flux is an important sink of nitrogen in the sediment of the Harima Nada, constituting about 30% of the sinking flux of PON into the sediment. The flux of N_2 estimated in our study is about one to two orders of magnitude smaller than the values reported in other areas such as the East China Sea and Arctic shelf. This discrepancy may be attributed to higher bacterial activity in those areas or to elevated concentrations of organic matter and NO_3^- in the sediment (Sun et al., 2021; Canion et al., 2014; Lin et al., 2017; Rich et al., 2020).

Our results indicate that the anammox rate is approximately three times greater than the denitrification rate. However, some previous studies reported that anammox contributes a smaller N_2 flux compared to denitrification (Teng and Lin, 2024; Dalsgaard et al., 2005). The first reason for this difference is the lower maximum NO_3^- concentration or shallower NO_3^- penetration in Harima Nada sediments, which leads to a denitrification rate lower than that in those regions. In addition, the low concentration of NH_4^+ in the sediment of those regions, even as the same scale as the NO_3^- concentration, limits the rate of anammox and reduces the contribution to N_2. In contrast, NH_4^+ concentration in the sediment in this study was significantly higher than NO_3^- concentration, thereby enhancing the potential rate of anammox (Canion et al., 2014; Rich et al., 2020). Another reason is that the sediment thickness utilized in our model was set at 10 cm, whereas most conventional observations sampled only the surface sediment, potentially underestimating the contribution of anammox in deeper layers. Usually, NH_4^+ concentrations in pore water are much lower in the surface layer compared to deeper layers, resulting in a reduced anammox reaction rate in the 0-5 cm layer relative to the 5–10 cm layer. When concentrating on the upper 5 cm of sediment, our model results suggest that the anammox rate constitutes a smaller

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proportion of N_2, accounting for less than 50% in summer (Figure S5 and Text S5), which is consistent with some observations (Teng and Lin, 2024; Dalsgaard et al., 2005).

5 Conclusions

This study developed a one-dimensional vertical sediment model to investigate the seasonal variations in the direction and magnitude of nitrogen fluxes at the sediment-water interface in semi-enclosed inland seas, and to reveal the controlling mechanisms of various environmental factors on these variations. Utilizing continuous observational data from a typical semi-enclosed inland sea for validation, we found that the flux of PON settling from the bottom water to the sediment is the main source of nitrogen in the sediment, with its seasonal variation governed by the PON concentration in the bottom water. Due to the regeneration of a substantial amount of NH_4^+ in the sediment, NH_4^+ is continuously released from the sediment to the overlying water, with its seasonal variation being temperature-controlled. Temperature influences the mineralization rate, which subsequently affects NH_4^+ flux through NH_4^+ inventory in the sediment. NO_3^- flux at the interface is directed upward in an oligotrophic water column, and its seasonal variation is also temperature-controlled. The influence of temperature over NH_4^+ inventory in the sediment is transmitted to NO_3^- inventory through nitrification, thereby affecting NO_3^- flux. This results in the seasonal variation of DIN flux returning from the sediment to the water being unrelated to the PON flux, as these two are controlled by different environmental factors with distinct seasonal patterns. This finding suggests that neglecting to consider bottom water temperature and PON concentration in reflective boundary conditions or empirical formulas may lead to varying degrees of misestimation when calculating the seasonal variation of DIN flux at the sediment-water interface.

The rate of nitrogen removal in sediments with low nitrate levels, possibly found in oligotrophic environments, is primarily controlled by anammox. Moreover, traditional observations are limited by the sampling depth of sediments, which may result in an underestimation of the contribution of anammox in deeper sediment layers. Our findings indicate that the maximum concentration of DO in sediments, which ranged from 60 to 140 mmol m^{-3}, has a negligible impact on sediment nitrogen cycling. However, seasonal variations in oxygen penetration depth, which ranges from 1 to 3 mm, can significantly influence nitrogen fluxes at the sediment-water interface. In addition, this model is mainly suited for semi-enclosed

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inland seas dominated by muddy sediments and does not account for the advection of pore water flow in sandy sediments. In practice, this process can be incorporated by adjusting the vertical velocity in the solute transport equation. Overall, this model can be widely applied to investigate nitrogen cycling-related issues in nearshore muddy sediments. Moreover, by investigating the seasonal variations of nitrogen cycling in semi-enclosed inland sea sediments and their controlling factors, it can provide a basis for governance and management support for environmental issues such as coastal eutrophication. In future research, we will expand upon this study by integrating it with long-term observational data to investigate the effects of prolonged changes in aquatic environments, such as eutrophication and seawater warming, on sediment nitrogen cycling and nitrogen flux at the sediment-water interface.

Code, data, or code and data availability

The source code of the numerical model used in this study is available on request. Please contact the corresponding author.

Author contributions

Zhaosen Wu: conceptualization, methodology, software, validation, visualization, writing (original draft preparation). Xinyu Guo: formal analysis, methodology, resources, supervision, writing (review and editing). Jie Shi: supervision, writing (review and editing). Xiaokun Ding: methodology, software. Masatoshi Nakakuni: data curation, investigation, resources. Kuninao Tada: data curation, investigation, resources.

Competing interests

The authors declare that they have no conflict of interest.

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Supporting Information

Supporting Information is available free of charge at the Biogeosciences website.

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