

## Response to Reviewer #1

Always given as follows:

Referee comment: RC; Author response: AR; Changes to the manuscript

### General comments

This is a numerical model study to identify the contribution of three sources of nitrogen, which is from the land, from the open sea, and from the seafloor to DIN in a semi-enclosed coastal sea (Seto Inland Sea: SIS). Although this information is important for the prevention of eutrophication in the sea, the calculation method has some problems as shown below.

Thank you for your valuable comments on this study. We have carefully considered your comments regarding the calculation method and would like to address them in this response note.

### Specific comments

**RC1:** Why do you include seafloor sources as a nitrogen source in addition to the open sea and terrestrial sources?

Unlike the case of phosphorus, nitrogen leached from the seafloor is the result of mineralization of “new” sediments, so nitrogen originating from the seafloor may be included in the open sea nitrogen and land nitrogen.

**AR1:** Thank you for your suggestion. We agree with your idea that the nitrogen leached from the seafloor is the result of the mineralization of “new” sediments. The nutrients contained in “new” sediments have an initial origin from either the rivers or the open ocean. The reason we treated it as a source is that many scientists always argue about the contribution of sediment to the nutrient in the water column. Therefore, this is just a practice way to define the nutrient source.

As we consider the sediment source of nutrients, we also realize that it has different processes from those from rivers and the open ocean. First, the processes by which particulate organic nitrogen (PON) settles onto the sediment, decomposes into dissolved inorganic nitrogen (DIN), and returns to the water column are different from the directly discharged nutrients of riverine or oceanic DIN, as well as the remineralization of PON in the water column. The sediment-released nutrients are suggested to be particularly important in shallow waters (Radtke et al., 2019). Second, there is a time lag between the deposition and mineralization of PON, either short-term or long-term (Soetaert et al., 2000). This means a large uncertainty in the proportion of nutrients released from sediment as compared to those supplied from rivers and the open ocean. Third, it is more difficult to control the nutrients released from the sediment than those supplied by rivers. Therefore, identifying the areas where the sediment-released nutrients dominate is helpful to the effective control or regulation of the riverine nutrients.

In conclusion, it is still necessary to track the sediment-released nutrients as a separate source. To address

your comment, we will mention that the nutrients leached from the seafloor have an origin from either rivers or the open ocean in Section 2.3. In addition, based on the proportion of PON flux settled on the sediment surface originating from the open ocean and rivers, we will give a quantitative estimation of the ratio of riverine and oceanic nutrients in the sediment-released nutrients in Section 4.2.

**Reference:**

Radtke, H., Lipka, M., Bunke, D., Morys, C., Woelfel, J., Cahill, B., Böttcher, M. E., Forster, S., Leipe, T., Rehder, G., and Neumann, T.: Ecological ReGional Ocean Model with vertically resolved sediments (ERGOM SED 1.0): coupling benthic and pelagic biogeochemistry of the south-western Baltic Sea, *Geosci. Model Dev.*, 12, 275–320, <https://doi.org/10.5194/gmd-12-275-2019>, 2019.

Soetaert, K., Middelburg, J. J., Herman, P. M. J., and Buis, K.: On the coupling of benthic and pelagic biogeochemical models, 29, 2000.

The sentences added to the revised manuscript are:

Added in Section 2.3:

“It needs to note that the nutrients in the sediment are originally mostly from the land, or the open ocean and the sediment is a temporary storage or a permanent sink. In this study, however, we treat the sediment as the third source to track. This is because the sediment-released nutrients are gaining more attention and particularly important in shallow waters (Radtke et al., 2019).”

Added in Section 4.2:

“If the proportion of PON flux settled on the sediment surface produced by the oceanic and riverine nutrients were taken as the proportion of DIN flux released from the sediment, among the  $86 \text{ mol N s}^{-1}$  of DIN from the sediment,  $60 \text{ mol N s}^{-1}$  has an origin from the open ocean and  $26 \text{ mol N s}^{-1}$  from rivers.”

Radtke, H., Lipka, M., Bunke, D., Morys, C., Woelfel, J., Cahill, B., Böttcher, M. E., Forster, S., Leipe, T.,

**RC2:** Why is dissolved organic nitrogen not included in the calculation?

Dissolved organic nitrogen, which accounts for about 90% of the total nitrogen in SIS, is not included in the calculation. The Ministry of the Environment's total load reduction for SIS is also based on total nitrogen in its calculations.

**AR2:** Thank you for your comment. Each biogeochemical model is designed for a different purpose, and the number and type of variables and biogeochemical processes are selected depending on the purpose (Fennel et al., 2022). The introduction of more variables and processes will introduce more uncertainties and calculation costs. In addition to expressing biogeochemical processes explicitly, expressing them implicitly is a substitute. The purpose of this study is to evaluate the inventories of nutrients and phytoplankton originating from different sources in the SIS. The direct transformations between them are of the most concern. Therefore, the widely used NPZD model including DIN, PHY, ZOO, and PON, is sufficient for the research purpose. For example, to estimate the nitrogen fluxes in the shelf area of the Middle Atlantic Bight, Fennel et al. (2006) also used an NPZD model to represent the nitrogen processes in the water column.

Although the research about the dissolved organic nitrogen (DON) in SIS was not often reported and the dataset of DON was also little, we still collected some results. In Hiuchi Nada and Iyo Nada, which are two sub-regions of SIS, Kumamoto et al. (1994) estimated the DON concentration at two sampling stations in late spring. They reported that DON concentration was superior to DIN concentration and nearly constant in the vertical and showed little temporal variation in Iyo Nada. By subtracting DIN from TN, Asahi et al. (2019) obtained the DON concentration at the surface layer of SIS in the 1990s and 2000s. They reported that DON concentration was higher than DIN and was relatively constant. Because we are interested in the materials that were dynamically changed, we did not include the little changed DON in our model. In other words, DON is like a background for TN but only DIN has a strong relationship with the low-trophic ecosystem. After reading your comment, however, we acknowledge that the incorporation of DON can be considered for some specific questions if there is a sufficient amount of observational DON to support model construction.

**Reference:**

Asahi T., Abo K., Abe K., and Tada K.: Comparison of Dissolved Inorganic and Organic Nitrogen between the sand s in the Seto Inland Sea of Japan, *Bulletin on Coastal Oceanography*, 56, 123–131, 2019.

Kumamoto, Y., Tsubota, H., and Fujiwara, K.: Temporal Variation of Dissolved Organic Nitrogen and Phosphorus, *Environmental Science*, 7, 1–12, <https://doi.org/10.11353/sesj1988.7.1>, 1994.

**RC3:** Are the DIN boundary conditions used in the numerical model reasonable?

The boundary condition is that the open sea origin DIN is zero at the seafloor and landward.

If there are no biochemical processes in SIS, no DIN supply from land or seafloor, only physical diffusion, then the DIN concentration in SIS is equal to the open boundary DIN (DIN from the open sea) and SIS is filled with DIN from the open sea. In other words,  $DIN = 0$  does not occur on the seafloor surface or the landward shore.

**AR3:** Yes, the DIN boundary conditions used in the numerical model and the tracking cases are reasonable. From the mathematical perspective, the numerical model is represented as a system of coupled partial differential equations. The tracking method we used in this study is a linear decomposition of the original partial differential equations. In order to solve the subset of partial differential equations, the boundary conditions also need to be linearly decomposed. For the boundary condition of the tracking open ocean case, we specified the open ocean origin DIN zero at the land boundary and seafloor. This is because the open ocean origin DIN is not released into the SIS from the land or seafloor. The case you said that SIS is filled with DIN from the open sea is the analytical solution of a diffusion equation with zero flux from land and a fixed concentration at the open ocean side. We actually used the same boundary conditions as the case you said. In our results, the open ocean-origin DIN is not zero at the landward shore or on the seafloor surface (Fig. 5b and Fig. 5e).

## Technical correction

**RC4:** It should be noted that the land load in this report is an underestimate.

The terrestrial nitrogen load for SIS is published every five years by the Ministry of the Environment of Japan (MEJ). It is necessary to state the values of the terrestrial load by MEJ and the terrestrial load in this report.

In SIS, which experienced eutrophication in the 1970s, the majority of domestic and industrial wastewater is treated at treatment facilities on the waterfront and discharged directly into the sea in recent years. Therefore, there is a large difference between the DIN flow via rivers and the total nitrogen flow actually entering the sea (especially in the eastern Seto Inland Sea).

In SIS, river discharge is significantly lower in winter, resulting in large seasonal variations in DIN flow from rivers, whereas there is little seasonal variation in DIN flow from domestic and industrial sources. This affects the seasonal variation of DIN concentration in the SIS.

**AR4:** Thank you for your suggestions. We have recognized that the DIN load from the land was underestimated in our study, and we mentioned it on Line 115 to Line 117. In the revised manuscript, we will appropriately increase the DIN load from the land using the total nitrogen (TN) load for SIS published every five years by the Ministry of the Environment of Japan, and we will give a quantitative estimate of how large this underestimation in the DIN load from land is. In this response letter, we first report the related information we have collected.

TN loads from land to the SIS were estimated by the Ministry of the Environment, Japan every five years from 1979 based on the unit load method in the catchment area of SIS (Abo and Yamamoto, 2019; Timita et al., 2016). From 1979 to 2014, the average TN load from land to the SIS was  $471 \text{ mol N s}^{-1}$ . Yanagi and Ishii (2004) indicated that the TN loads estimated by the unit load method did not reproduce the inflow of TN to the coastal sea since some parts of TN loads remained on land. Yamamoto et al. (1996) recommended the use of river flow rate to calculate the actual inflow TN load into the SIS and reported that the TN load calculated using this method was about 48% of that measured by the unit load method. Based on this value presented by Yamamoto et al. (1996), the average inflow TN load to the SIS was  $226 \text{ mol N s}^{-1}$ , which flowed into the SIS from the land through the 21 first-order rivers and about 640 other small rivers. We included 21 first-order rivers and 45 small rivers in our study. In addition, the compounds of TN loads from land are not clear. The proportion of DIN concentration in TN concentration at the first-order rivers of SIS is about 77%, which was estimated by the nutrient data from the Ministry of Land, Infrastructure, Transport and Tourism, Japan (<http://www1.river.go.jp/>). If we apply this value to the TN load from land, the DIN load from land is  $174 \text{ mol N s}^{-1}$ , which is 2.7 times higher than the DIN load from rivers ( $64 \text{ mol N s}^{-1}$ ) estimated by our study. A more extreme situation is that in addition to DIN, other compounds of TN can also be used by the phytoplankton through complex biogeochemical processes. Then our estimation of  $64 \text{ mol N s}^{-1}$  will be 28% of the TN load from land. In order to consider the DIN load from land as much as possible in revision, a new series of experiments is conducted to increase the DIN load from the rivers to 3 times the original value ( $64 \text{ mol N s}^{-1}$ ) to represent the DIN load from land. These experiments are being calculated and the new results

will be described in the revised manuscript.

**Reference:**

Abo, K. and Yamamoto, T.: Oligotrophication and its measures in the Seto Inland Sea, Japan, *Bulletin of Japan Fisheries Research and Education Agency*, 49, 21–26, 2019.

Tomita, A., Nakura, Y., and Ishikawa, T.: New direction for environmental water management, *Marine Pollution Bulletin*, 102, 323–328, <https://doi.org/10.1016/j.marpolbul.2015.07.068>, 2016.

Yamamoto, T., Kitamura, T., and Matsuda, O.: Riverine inputs of fresh water, total nitrogen and total phosphorus into the Seto Inland Sea, *Journal of the Faculty of Applied Biological Science, Hiroshima University*, 35, 81–104, 1996.

Yanagi, T. and Ishii, D.: Open Ocean Originated Phosphorus and Nitrogen in the Seto Inland Sea, Japan, *J Oceanogr*, 60, 1001–1005, <https://doi.org/10.1007/s10872-005-0008-4>, 2004.

**RC5:** Section 3.2. It is important to indicate the time required for the numerical model to become stationary; the DIN flow path during the set-up period is not the flow path when the model becomes stationary.

**AR5:** Thank you for your suggestions. We indicated the time required for the numerical model to become stationary in Section 2.2. In Lines 150 to 151: “The hydrodynamic-biogeochemical model was initiated on the first day of January and stabilized from the third year onwards. Therefore, the simulation results of the third year were used to analyze the seasonal variations of DIN and PHY.” For the tracking case, at Line 171: “We initiated the tracking technique from the first day of the fourth year of the hydrodynamic-biogeochemical model, ...” and at Line 178 to 179: “After a spin-up of three years, the annual cycle of each source of nutrients and related particles became stationary.” In order to make readers clear about the time required for the numerical model to become stationary, we will add one figure to describe the ratio of  $(\text{DIN}_{\text{ocean}} + \text{DIN}_{\text{river}} + \text{DIN}_{\text{sediment}}) / \text{DIN}$  in the SIS from the first year to the third year of the tracking simulation in the revised Supplement Materials.

The purpose of Section 3.2 is to exhibit the pathway of oceanic, riverine, and benthic DIN gradually occupying the SIS from the initial state. After one year of calculation, DIN concentrations from the open ocean, rivers, and sediment have already occupied most areas of the SIS. Therefore, we presented the results of the first year of tracking cases in Fig. 4 to depict this pathway.

**RC6:** Actual measurements of the amount of nitrogen and phosphorus entering SIS from the open sea were made by several organizations in the 1980s to 2000s, and it has been shown that the amount of nitrogen entering from the open sea is equivalent to the amount of land-based load during the summer months. It is desirable to cite these papers.

**AR6:** Thank you for your suggestions. In revision, we collected the related papers and present them in this response note. We will cite these papers in Section 4.2 of the revised manuscript.

Fujiwara et al. (1997) reported that about  $28 \text{ mol s}^{-1}$  of TN was transported from the open ocean to the SIS

through Bungo Channel based on observations during 15 days from July to August in 1982 and  $140 \text{ mol s}^{-1}$  of TN through Kii Channel from open ocean based on observations during 2 days in August of 1985. Kasai et al. (2001) estimated the net DIN transport through Kii Channel into SIS in August of 1996 was  $111 \text{ mol s}^{-1}$  which was similar to that of Fujiwara et al. (1997). However, they also report that in Augusts of 1997 and 1999, only  $6.5 \text{ mol s}^{-1}$  of DIN and  $37 \text{ mol s}^{-1}$  of DIN were transported into the SIS, respectively.

By subtracting the amount of DIN load buried in the sediment from the amount of DIN load from land, Fujiwara et al. (2006) reported that  $50 \text{ mol s}^{-1}$  of TN was transported from the open ocean to the SIS.

Although quantitative estimates of DIN transport from the open ocean were not available, Takashi et al. (2002) revealed a strong (weak) inflow of DIN transport to the SIS when the Kuroshio is offshore (inshore) in summer, but the outflow in winter was regardless of the path of Kuroshio based on monthly observations from April 1999 to December 2001.

#### Reference:

Fujiwara, T., Uno, N., Tada, M., Nakatsuji, K., Kasai, A., and Sakamoto, W.: Inflow of nitrogen and phosphorus from the ocean into the Seto Inland Sea, Proc. Coastal Engineering (JSCE), <https://doi.org/10.2208/proce1989.44.1061>, 1997.

Fujiwara, T., Kobayashi, S., Kunii, M., and Uno, N.: Nitrogen and phosphorus in Seto Inland Sea: Their origin, budget and variability, Bull Coast Oceanogr, 43, 129–136, 2006.

Kasai, A., Fujiwara, T., and Tada, M.: Ocean Structure and Nutrient Transport in the Kii Channel, Proceedings of Coastal Engineering, 48, 436–440, <https://doi.org/10.2208/proce1989.48.436>, 2001.

Takashi, T., Fujiwara, T., Sumitomo, T., and Takeuchi, J.: Transport of nitrogen and phosphorus from the open ocean to the Kii Channel, Proceedings of Coastal Engineering, 49, 1076–1080, <https://doi.org/10.2208/proce1989.49.1076>, 2002.

**RC7:** Line 114: It should be noted that the seasonal variation of the nitrogen load from rivers is due to the seasonal variation of the river flow. Unlike Europe, the SIS receives a little precipitation in winter.

**AR7:** Thank you for your suggestions. We will note this information in the revised manuscript.

The revised expression is:

“The seasonal variation in DIN loads of all rivers, with high loads in July and September and low loads in January, is primarily controlled by the seasonal variation in river discharge. The annual mean of DIN loads from rivers is  $63.85 \text{ mol s}^{-1}$ .”