

Review

Tian et al. used the isotope mass balance model to characterize and quantify reactive nitrogen sources and sinks in the Bohai Sea through the measurements of nutrient, nitrate $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$, and $\delta^{15}\text{N}$ of suspended matters and sediments. The authors used the results both from their work and previous studies trying to give a more comprehensive estimation of nitrate sources and sinks from various end members. This work would improve the understanding of N cycle in the Bohai Sea, a typical semi-enclosed bay influenced by anthropogenic nitrogen input, I think this manuscript could be accepted after a minor revision. Here, I have some specific comments for this study:

We thank the reviewer for a positive and encouraging review and thoughtful comments and queries. In the following we address the points raised and how they will be implemented in the revision.

1. I think this study would need a little bit more detail discussing of the model uncertainties. There could be some uncertainties in this isotope mass balance mode due to many assumptions in this study. For example, there are many assumptions for using the end member of sedimentation (section 4.2.5). As the isotope fractionation associated with the processes of assimilation and nitrification is complicated, I think it may not be suitable to give fixed values of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ to the correlated end members. I suggest to give varying values of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ with reasonable range when applying to the isotope mass balance mode.

Authors' reply: The reviewer is of course correct in pointing out the errors arising from adopting fixed end member values. We will set up the uncertainties for the end members in the revised manuscript.

2. In addition, in summer, nitrate was almost depleted in the most area of the Bohai Sea, suggesting an enhanced photosynthesis rate and assimilation rate in this season. The residual nitrate would have high $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ values. It may need to evaluate rationality by adopting average values of nitrate concentrations, $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ in the two seasons when applying to the isotope mass balance model.

Authors' reply: Thanks for this suggestion that we will implement in the revision (see supplement below). As mentioned in the manuscript, only a subset of samples could be analyzed due to the low nitrate concentrations in summer, and most of these are from the Yellow River Diluted Water that had $[\text{NO}_3^-] > 1.7 \mu\text{mol/L}$. The average values of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ of the Bohai Sea in summer were $9.9 \pm 3.5\%$ (n=23) and $8.7 \pm 3.3\%$ (n=23). Although measurements could not constrain the range of nitrate isotope values, the lacking isotope data can be roughly estimated:

According to the T-S patten in summer, the Bohai Sea water can be considered as a two-end member mixture between fresh water discharged from Yellow River (YR) and sea water of central Bohai Sea, the nitrate concentration only affected by physically mixing hence can be calculated (Supplement 1). The isotope effect of assimilation for nitrate in the Bohai Sea follows the “steady-state model” rather than the Rayleigh model because the Yellow River supplies nitrate continuously (Sigman and Fripiat, 2019). Thus, the estimated dual nitrate isotope values can be calculated according to equation (1) and (2):

$$\delta^{15}\text{N}_{\text{reactant}} = \delta^{15}\text{N}_{\text{initial}} + {}^{15}\epsilon(1 - f) \quad (1)$$

$$\delta^{18}\text{O}_{\text{reactant}} = \delta^{18}\text{O}_{\text{initial}} + {}^{18}\epsilon(1 - f) \quad (2)$$

In Eq.1 and Eq. 2, f is equal to the observed nitrate concentration divided by of result of the two-

end member model, $\delta^{15}\text{N}_{\text{initial}}$ is equal to the end member of YR, and $\delta^{15}\text{N}_{\text{reactant}}$ is the estimated value of the residual nitrate, the value we need. The average of $^{15}\epsilon$ and $^{18}\epsilon$ adopted here are 5‰ (Granger et al., 2010; DiFiore et al., 2009; Liu et al., 2017; Wu et al., 2019; Umezawa et al., 2013; Wang et al., 2016).

The readjusted values of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ for the Bohai Sea in summer is $12.8 \pm 2.7\text{‰}$ (n=85) and $9.1 \pm 1.9\text{‰}$ (n=85), respectively, resulting in seasonally averaged values of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ of 10.3‰ and 10.6‰, respectively. These values induce about 5%-9% deviations of the mass fluxes in our box model. Because this estimate is also based on the two-end member mixing model and isotopic fractionation equations, we think that this part probably is better placed in the uncertainty discussion that will be included in the revision.

Supplement 1 The estimate of two end member mixing of nitrate

The YR provides warm, fresh and nitrate enriched water whereas cold, saline and nitrate depleted water was observed near the area of the outer Liaodong Bay in both spring and summer. Thus, there were two end members to be considered in a mixing model. One should be aware that a contribution of atmospheric nitrogen is included in the marine end member as well.

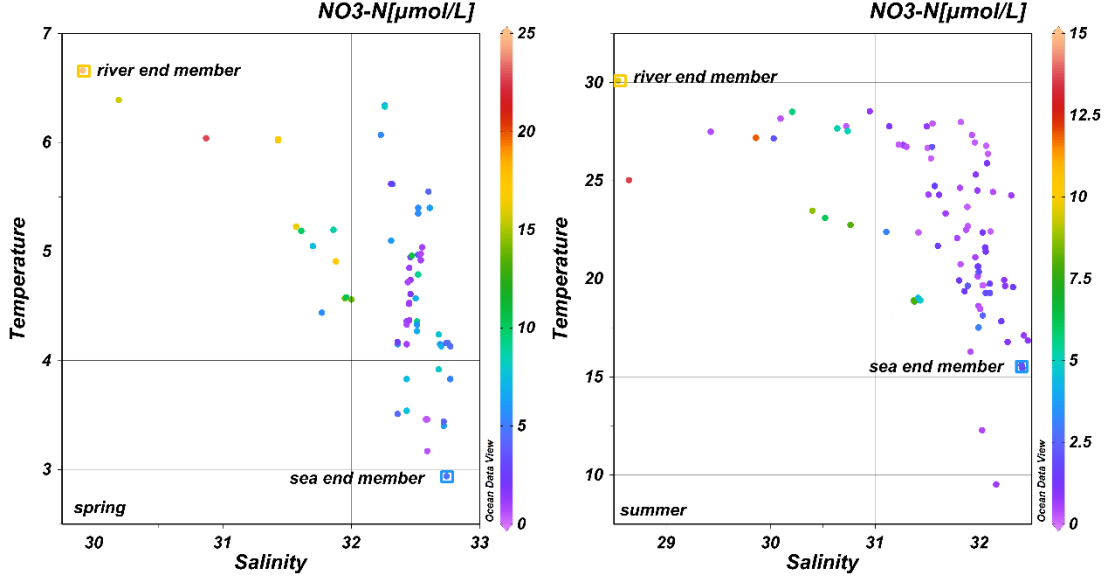


Fig. S1 Temperature vs. salinity in Bohai Sea in spring (left) and summer (right). The values adopted for the two nitrate end members were mainly based on this pattern

The values of these two end members are shown in Table S-1. The summer basic pattern of temperature and salinity was similar to that of spring. Thus, the fraction of water originating from YR and the BHS during the mixing process can be calculated follow (1) and (2):

$$S = S_r \times f_r + S_s \times f_s \quad (1)$$

$$f_r + f_s = 1 \quad (2)$$

where S , S_r and S_s refers to the observed salinity in study area, the end member value of river and sea, respectively. f_r and f_s refers to the fraction of river and sea water, respectively. The modeled nitrate concentration and modeled $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ values can be calculated following equations (3), (4) and (5):

$$[\text{NO}_3^-]_m = [\text{NO}_3^-]_r \times f_r + [\text{NO}_3^-]_s \times f_s \quad (3)$$

$$\delta^{15}\text{N}_m[\text{NO}_3^-]_m = \delta^{15}\text{N}_r[\text{NO}_3^-]_r + \delta^{15}\text{N}_s[\text{NO}_3^-]_s \quad (4)$$

$$\delta^{18}\text{O}_m[\text{NO}_3^-]_m = \delta^{18}\text{O}_r[\text{NO}_3^-]_r + \delta^{18}\text{O}_s[\text{NO}_3^-]_s \quad (5)$$

where $[\text{NO}_3^-]_m$, $[\text{NO}_3^-]_r$ and $[\text{NO}_3^-]_s$ refers to the modeled nitrate concentration and the end member nitrate concentration values of river and sea, respectively. $\delta^{15}\text{N}_m/\delta^{18}\text{O}_m$, $\delta^{15}\text{N}_r/\delta^{18}\text{O}_r$ and $\delta^{15}\text{N}_s/\delta^{18}\text{O}_s$ refer to the modeled $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ values, and the end member $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ values of river and sea, respectively.

Table S-1 Two end member values in Bohai Sea

		Riverine	Marine
spring	Salinity	29.9	33.0
	Nitrate/ $\mu\text{mol/L}$	31.1	6.0
	$\delta^{15}\text{N}\text{‰}$	9.5	6.0
	$\delta^{18}\text{O}\text{‰}$	6.8	12.5
summer	Salinity	28.5	32.5
	Nitrate/ $\mu\text{mol/L}$	13.6	2.0
	$\delta^{15}\text{N}\text{‰}$	9.9	9.5
	$\delta^{18}\text{O}\text{‰}$	5.3	8.2

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