Point-by-point response to the reviewers' comments

The comments from the reviewers are in bold followed by our responses in regular text. The text in quotation marks represents the content we revised in the new manuscript.

Major comments

<u>Reviewer 2 – Major remark 4</u>

It seems that you misunderstood my comment. Maybe I should have phrased my comment clearer. I was not talking about trends within a 10-years time period, but about that using only a ten-years mean as a reference may be misleading when comparing two 10-year means to determine the differences (i.e. trend) between these two climates. In hydrology, there is substantial decadal variability with wet and dry decades. Hence, if you accidentally compare a wet decade with a dry decade, you will find a difference (i.e. trend) that is not real just because of decadal variability. Usually, to investigate climatological relevant differences in hydrology, you have to compare at least 30-year means to get robust results.

Thank you for your detailed explanation. According to your suggestion and considering the rapid increase of nitrogen (N) fluxes since 1960, we used the 20-year average N fluxes of 1901-1920 and 1995-2014 to quantify the changes from early 20th century to the contemporary period (Figs 8, S10 b). We also analysed the contemporary spatial patterns of water discharges, N fluxes and N concentrations using a 20-years average from 1995 to 2014 (Figs 7, 10, S10 a, S11, and S12). With reference to the IPCC visual guide, we have adjusted the color scheme of the figures above to make them more friendly to color-blind individuals. The relevant content has been updated in the manuscript, mainly in section 3.2 "Temporal and spatial patterns of N flows".

“Averaged over the 1995-2014 period, the annual TN input from soils to rivers, TN exports to oceans and denitrification in transit amount to 64.4 Tg N yr^{-1}, 40.0 Tg N yr^{-1}, and 24.4 Tg N yr^{-1}, respectively. These three N fluxes show increasing trends from 1901 to 2014. The global annual TN input to rivers increased by 72.4 %, from 37.4 Tg N yr^{-1} during 1901-1920 to 64.4 Tg N yr^{-1} during 1995-2014 (Fig. 6 a). The global annual TN export to oceans increased by 45.6 % from 27.4 Tg N yr^{-1} to 40.0 Tg N yr^{-1}. Most of this increase is attributed to DIN, which doubled over the simulation period, rising from 10.0 Tg N yr^{-1} to 19.9 Tg N yr^{-1}, while, in absolute terms, DON exports show a much smaller increase but still substantial relative increase of 50.6 % (Fig. 6b). In contrast, PON exports to oceans show a slightly decreasing trend. This decrease is mainly attributed to global greening, which enhances vegetation cover (Cortés et al., 2021; Wang et al., 2022) and reduces soil erosion, resulting in lower PON inputs from the land and, thus, PON exports to oceans. The increase in global denitrification mostly follows the rise in DIN inputs, with a relative increase of 146.6 %, from 9.9 Tg N yr^{-1} during 1901-1920 to 24.4 Tg N yr^{-1} during 1995-2014 (Fig. 6a).” (lines 601-617)

“

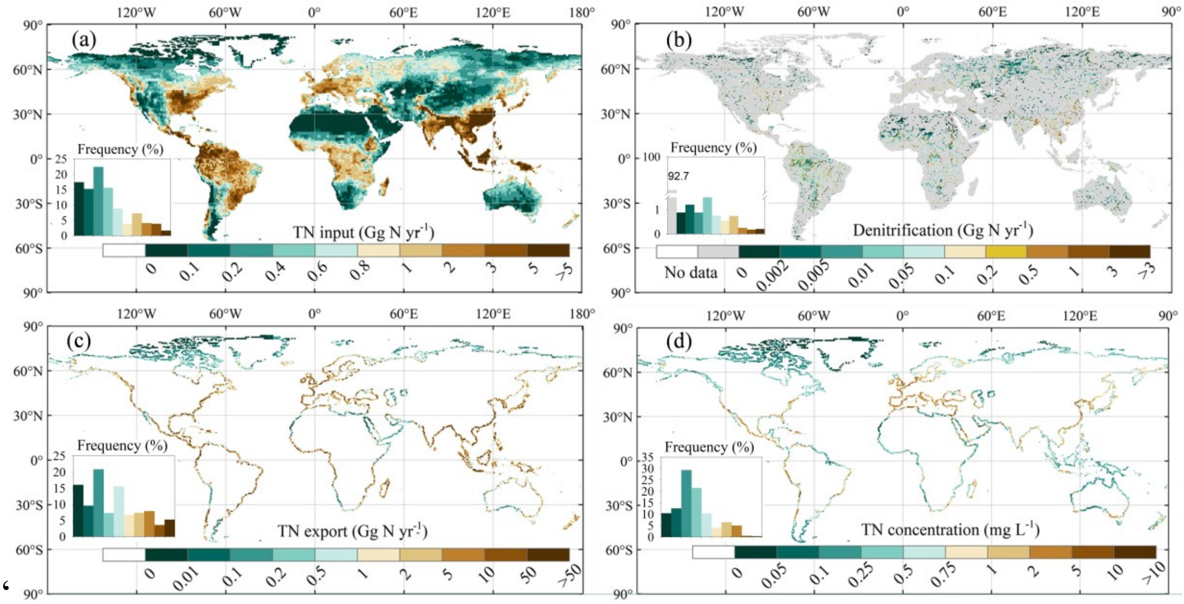

Figure 7. Spatial patterns of annual mean N fluxes and concentrations during 1995-2014: (a) TN inputs into rivers; (b) denitrification rates in rivers; (c) TN exports to oceans; (d) TN concentrations at rivers mouths. To display the spatial

patterns of denitrification in rivers better, we excluded data with denitrification rates less than 0.001 GN yr^{-1} per grid.” (lines 678-682)

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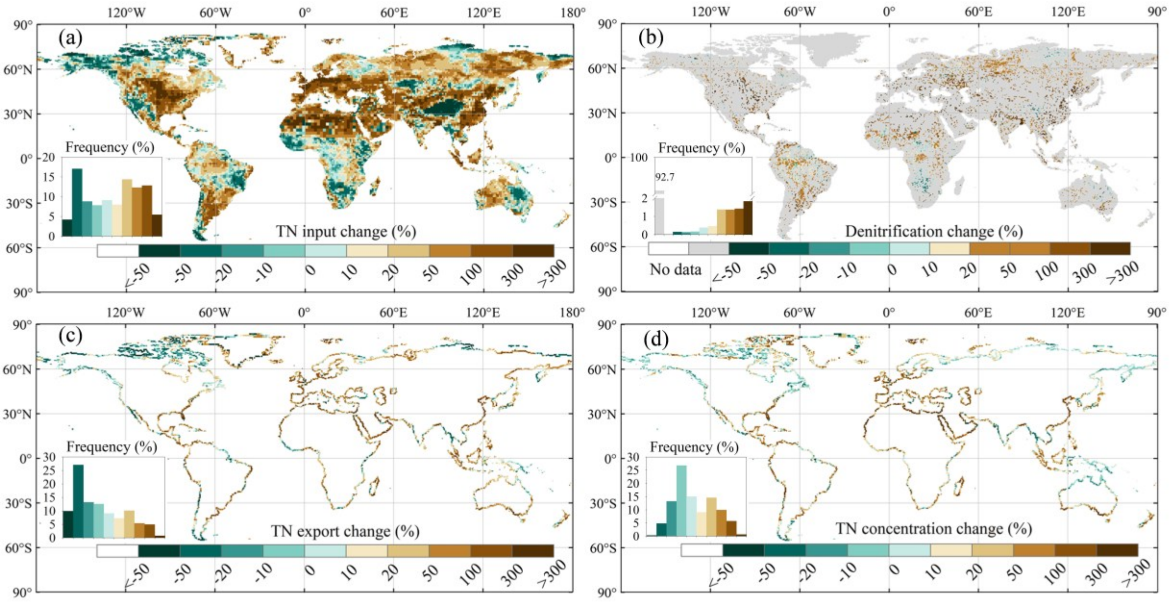

Figure 8. Spatial patterns of changes from 1901-1920 to 1995-2014 of: (a) TN inputs into rivers; (b) denitrification; (c) TN exports to oceans; (d) TN concentrations.” (lines 683-686)

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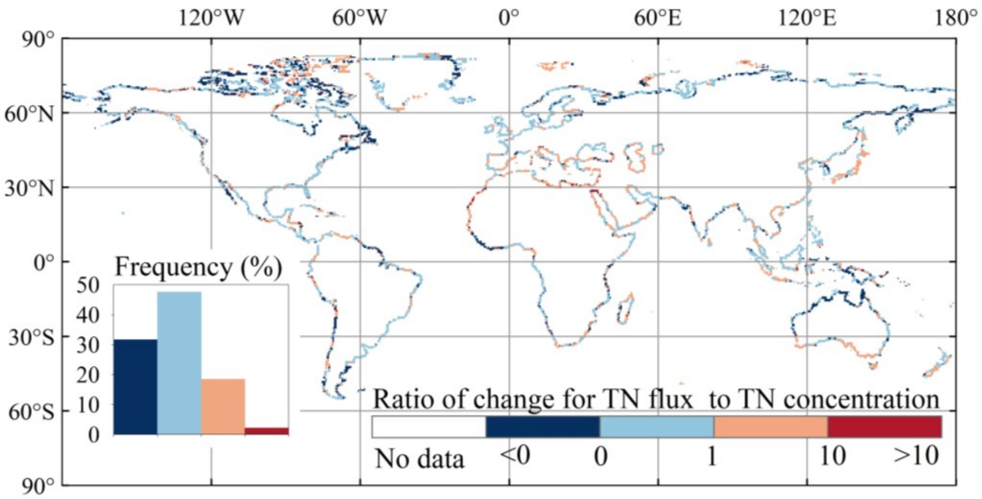


Figure 9. Ratio of changes in TN exports to changes in TN concentrations from 1901-1920 to 1995-2014.” (lines 687-689)

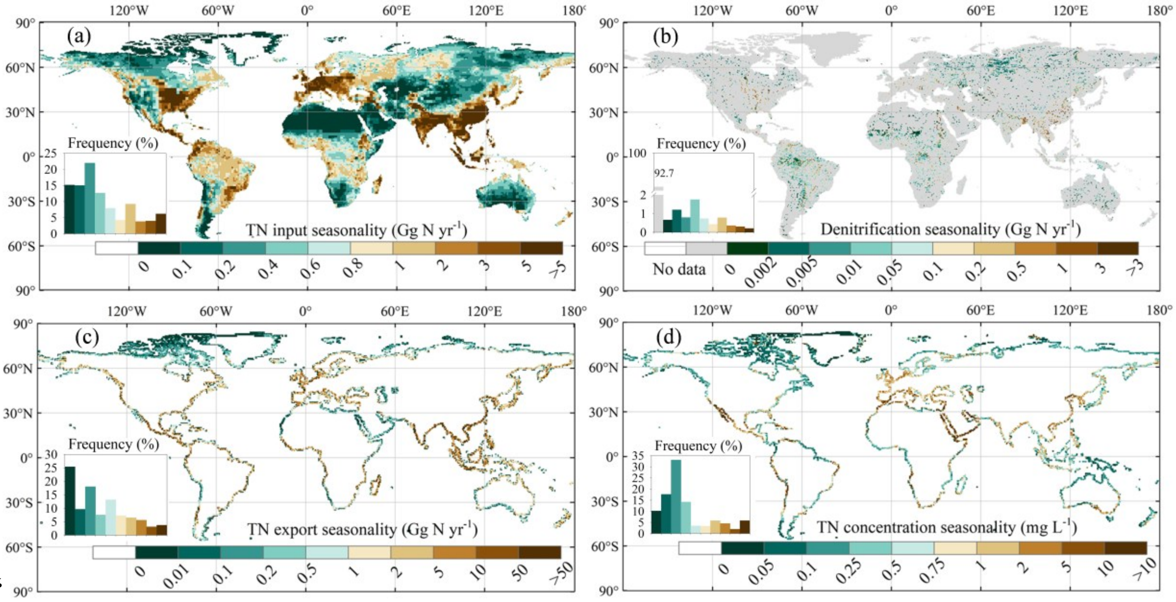
“ 

Figure 10. Spatial distribution of the seasonal amplitude (period 1995-2014) in: (a) TN inputs into rivers; (b) rates of denitrification; (c) TN exports to oceans; (d) TN concentrations at rivers mouths.” (lines 721-724)

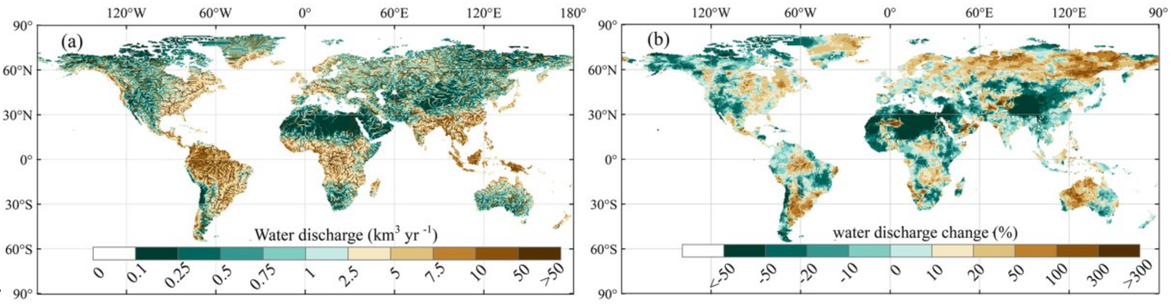
“ 

Figure S10. Spatial patterns of water discharge: (a) average annual water discharge over 1995-2014; (b) water discharge changes from the reference period 1901-1920 to 1995-2014.” (lines 50-53 in supplement)

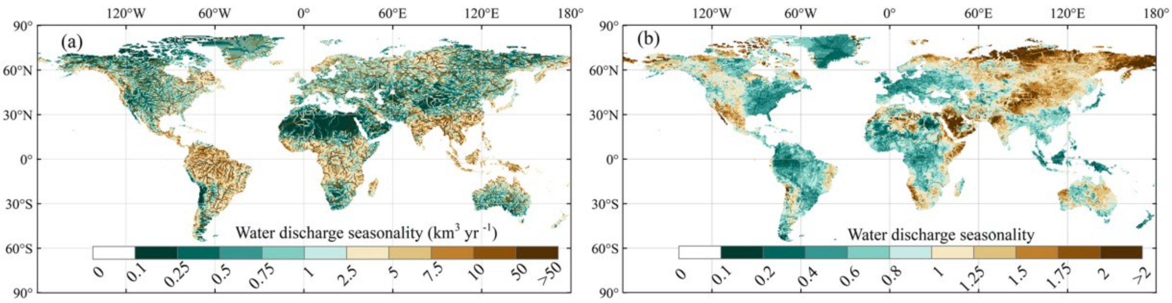
“ 

Figure S11. Spatial patterns of water discharge seasonality over 1995-2014: (a) water discharge seasonality; (b) normalized water discharge seasonality (=water discharge seasonality/ averaged annual water discharge).” (lines 54-57 in supplement)

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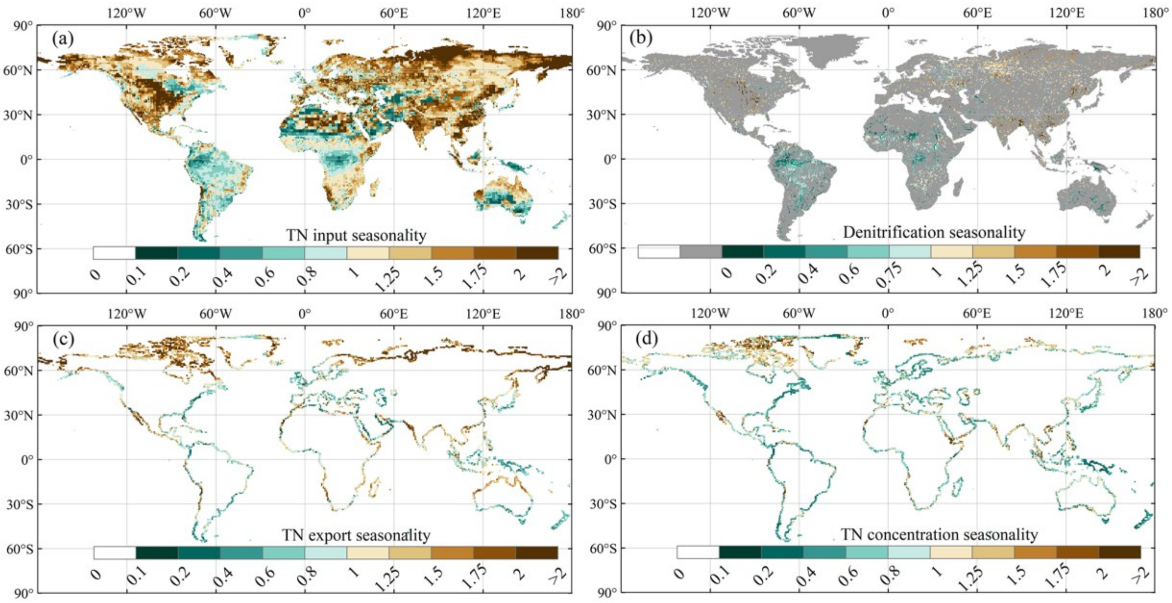


Figure S12. Spatial distribution of normalized seasonality for TN and denitrification over 1995-2014: (a) TN inputs into rivers; (b) denitrification rates; (c) TN export to oceans; (d)TN concentrations at rivers’ mouths. The normalized seasonality of TN or denitrification = seasonality of TN or denitrification /averaged annual values of TN or denitrification.” (lines 58-63 in supplement)

- **Minor comments**

1. **<u>Sect. 2.1.2</u>. Please note the meteorological forcing data (i.e. GSWP3) and the time period that has been used to simulate the runoff and drainage data. This is relevant information the reader should be able to get directly from the text without searching for it in Table 1.**

    Thanks for the great suggestion. We have added information of the meteorological forcing, land cover and soil parameters in the text.

    "ORCHIDEE-CNP and ORCHIDEE-Clateral used the same scheme to simulate soil hydrology (Sun et al., 2021; Zhang et al., 2022), and they have been run with the same climate forcing data, land cover map and soil parameters maps (Table 1). The climate forcing data during 1901-2014 were obtained from Global Soil Wetness Project Phase 3 (GSWP 3). Both ORCHIDEE-CNP and ORCHIDEE-Clateral used the ESA-CCI LUH2v2 plant functional type (PFT) distribution, which combines the ESA-CCI land cover map for 2015 with the historical land cover reconstruction from LUH2 (Lurton et al., 2020). Soil parameters in these two models follow Reynolds et al. (1999) and the Harmonized World Soil Database (FAO/IIASA/ISRIC/ISS-CAS/JRC, 2012)." (lines 211-220)

2. **<u>Line 209-211</u>.**

    ~~As~~ ORCHIEE-CNP … …2022), *and* they …

    Thanks, we have corrected it to:

    "ORCHIDEE-CNP and ORCHIDEE-Clateral used the same scheme to simulate soil hydrology (Sun et al., 2021; Zhang et al., 2022), and they have been run with the same climate forcing data, land cover map and soil parameters maps (Table 1)." (lines 211-214)

3. **<u>Line 212-214</u>.**

    Therefore, the *differences* in … …are *relatively* small.

    Thanks, we have corrected it.

"Therefore, the differences in runoff (0.9%) and drainage (1.7%) simulated by the two ORCHIDEE branches are relatively small (Fig. S1)." (lines 220-222)

4. **Line 254.**

… temporal *resolutions* of …

Thanks, we have corrected it to:

"S_{res} and T_{res} are the original spatial and temporal resolutions of the forcing data, respectively." (lines 262-263)

5. **Please be more thorough with shortenting unnecessary repetitions. Eq. 2-6 should still be merged into one equation, the same applies to eq. 10-12, and eq. 13-14**

Thank you very much for your advice. The Eqs. 2-8, combined with Fig. 2, clearly illustrate the processes of N transformation and transport. Although formulas 2-6 appear similar, we believe that this presentation improves readability. This demonstrates that the N lateral transfer process is represented through eight distinct N pools, fast PON reservoir, fast DON reservoir, fast DIN reservoir, slow DON reservoir, slow DIN reservoir, stream PON reservoir, stream DON reservoir and stream DIN reservoir. Therefore, we hope to retain Eqs. 2-6.

We have shortened original Eqs. 10-12 from

"$$F_{out\_PON} = S_{PON} \times \frac{F_{out\_H2O}}{S_{H2O}} \quad (10)$$

$$F_{out\_DON} = S_{DON} \times \frac{F_{out\_H2O}}{S_{H2O}} \quad (11)$$

$$F_{out\_DIN} = S_{DIN} \times \frac{F_{out\_H2O}}{S_{H2O}} \quad (12)$$

where all S terms represent N stocks (g N) and water stocks (m^3), and F terms represent flow rates of water (m^3 d^{-1}) and N (g N d^{-1}). F_{out_PON} represents PON flow rates from fast ($F_{fastout\_PON}$)/ stream ($F_{streamout\_PON}$) reservoirs; F_{out_DON} represents DON flow rates from fast ($F_{fastout\_DON}$)/ slow

($F_{slowout\_DON}$)/ stream ($F_{streamout\_DON}$) reservoirs; F_{out_DON} represents DIN flow rates from fast ($F_{fastout\_DIN}$)/ slow ($F_{slowout\_DIN}$)/ stream ($F_{streamout\_DIN}$) reservoirs. The same principle applies to the *S* (stocks) terms."

to

"$F_{out\_N} = S_N \times \frac{F_{out\_H2O}}{S_{H2O}}$ (10)

Where S_{H2O} represents water stocks (m^3), and F_{H2O} represents rates of water discharge (m^3 d^{-1}). F_{out_N} represents PON flow rates from fast ($F_{fastout\_PON}$) / stream ($F_{streamout\_PON}$) reservoirs, DON flow rates from fast ($F_{fastout\_DON}$) / slow ($F_{slowout\_DON}$) / stream ($F_{streamout\_DON}$) reservoirs, DIN flow rates from fast ($F_{fastout\_DIN}$) / slow ($F_{slowout\_DIN}$) / stream ($F_{streamout\_DIN}$) reservoirs. The same principle applies to the S_N (N stocks) terms." (lines 341-347)

And the original Eqs. 13-14 have been changed from:

"$\mathrm{R_{PON}} = \mathrm{S_{PON}} \times \mathrm{K_{PON}} \times \mathrm{Q10}^{\frac{\mathrm{TW}-\mathrm{T_{ref1}}}{10}}$ (13)

$\mathrm{R_{DON}} = \mathrm{S_{DON}} \times \mathrm{K_{DON}} \times \mathrm{Q10}^{\frac{\mathrm{TW}-\mathrm{T_{ref1}}}{10}}$ (14)

K_{PON} (0.028 d^{-1}) represents the average PON decomposition rate at 20°C in water (Islam et al., 2012); K_{DON} (0.07 d^{-1}) represents the average DON decomposition rate at the reference temperature of 20°C in water (Xia et al., 2013)."

to

"$R_{ON} = S_{ON} \times K_{ON} \times Q10^{\frac{TW-T_{ref1}}{10}}$ (11)

R_{ON} (g N d^{-1}) represents decomposition rate of organic N (ON, i.e., PON and DON); S_{ON} (g N) represents ON stocks in each reservoir. K_{ON} represents the average PON decomposition rate (K_{PON} = 0.028 d^{-1}) (Islam et al., 2012), and the average DON decomposition rate (K_{DON} = 0.07 d^{-1}) at the reference temperature of 20°C in water (Xia et al., 2013)." (lines 353-358)

6. **<u>Line 380</u>**

   ***Tables* A1 and A2 *provide* a …**

   Thank you for the suggestion. We have corrected it to:

   "Tables A1 and A2 provide a summary of all variables, fluxes and processes incorporated in LSM_Nlateral_Off." (lines 385-386)

7. **<u>Line 516-517</u>.**

   … water *discharge* (Fig. S3b).

   Thank you for the suggestion. We have corrected it to:

   "LSM_Nlateral_Off significantly underestimated (MBE < -100%) or overestimated (MBE > 100%) the observed TN flows at 32 sites (17% of all sites), all located in regions with relatively low water discharge (Fig. S3b)." (lines 522-525)

8. **<u>Line 589.</u>**

   In *the following*, we …

   Thanks, we have corrected it to:

   "In the following, we investigate spatial, seasonal and decadal trends resulting from the offline coupling of these three models." (lines 597-599)

9. **<u>Line 693-700</u>**

   It is written: "Our results indicate that the spatial pattern of seasonal amplitudes in TN concentrations at river mouths differs from that of TN exports (Fig. 10c, d). This result is important because the ocean biogeochemical modelling community typically uses annual mean TN fluxes derived from Global News to force their simulations (e.g., …), and downscales these inputs to monthly values under the assumption that the seasonal variability of the flux is entirely driven by river discharge."

   This statement is not very clear to me. Using the approach described, the community assumes constant TN concentrations CN that are applied to calculate TN exports at the river mouth by multiplying the river

discharge Q by CN. However, then the seasonal amplitude of CN (constant= zero amplitude) is also different to the one of the TN exports (seasonal amplitude induced by the discharge). In my opinion, to have a valid criticism of the common community method, you have to show that the seasonal amplitude of TN exports is significantly different to the seasonal amplitude of river discharge.

Apologies for any unclear expressions in section "2.2 Observational data." For observational data, we calculated the monthly total nitrogen (TN) flow by multiplying the monthly averaged TN concentrations by the monthly water discharge (Q). We didn't assume that TN concentrations remain constant throughout the year, meaning that seasonal amplitude of TN concentrations is not zero. We have added relevant explanation in section 2.2.

"We first calculated the monthly average N concentrations and monthly total water discharge, then determined the monthly N fluxes using Eq. 16. The total annual N flux is then obtained by summing the monthly N fluxes over the entire year." (lines 411-414)

It should be noted that the seasonal amplitude of TN exports and TN concentrations depicted in Fig. 10 are based on LSM_Nlateral_Off simulations rather than observations. The LSM_Nlateral_Off model operates with a daily time step, allowing for the calculation of daily TN concentrations and fluxes. From these daily results, monthly average TN concentrations and fluxes were derived, and the seasonal amplitude of TN concentrations and fluxes was quantified based on Eqs. 20-21. In order to provide visual representation of the differences between seasonality for TN fluxes and TN concentrations, we presented the ratio of normalized seasonality for TN fluxes to TN concentrations in Fig. S13.

"Our results indicate that the spatial pattern of seasonal amplitudes in TN concentrations at river mouths differs from that of TN exports (Figs. 10, S12, S13)." (lines 704-706)

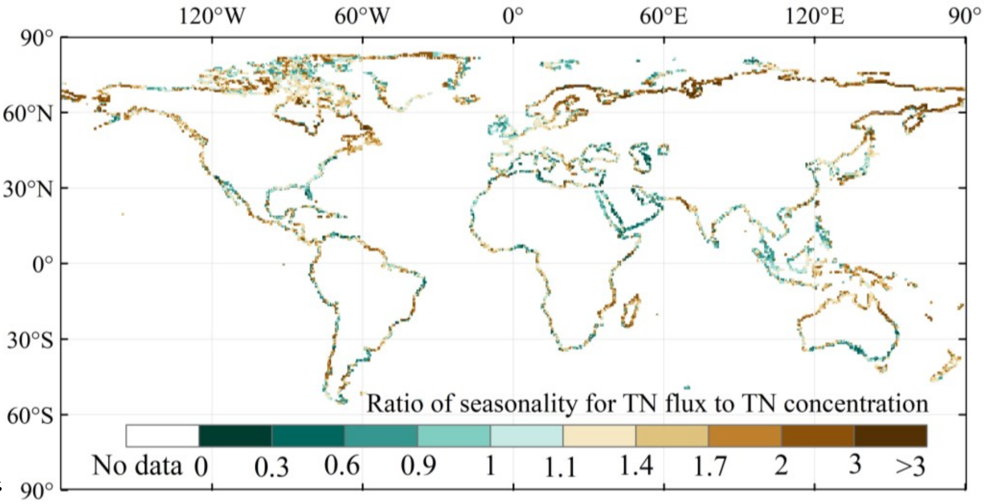
"


Figure S13. Spatial distribution of the ratio between normalized seasonality of TN flux and TN concentration during 1995-2014." (lines 64-66 in supplement)

10.Line 712

… (b) *rates of denitrification*; (c) *TN exports to oceans*; …

Sorry for the mistake, we have modified it to:

"Figure 10. Spatial distribution of the seasonal amplitude (period 1995-2014) in: (a) TN inputs into rivers; (b) rates of denitrification; (c) TN exports to oceans; (d) TN concentrations at rivers mouths." (lines 722-724)