### **Response to the reviewer #2**

We greatly appreciate the reviewer's comments and suggestions. We have made a concerted (and hopefully successful) attempt to address the reviewer's concerns in the revised manuscript.

First, we agree that PON exports from land to rivers should be assumed to occur with eroded soil fluxes. We have thus implemented this suggested approach and have made associated modifications throughout the manuscript.

Second, we agree that the discussion of the uncertainties in modeling soil erosion and associated fluxes to rivers and the ocean, as well as the discussion of uncertainties generally, was not sufficient in the previous manuscript. We have thus substantially extended the discussion and highlighted areas for priority future development.

Third, we agree that evaluation of the simulated algae dynamics was not sufficient in the previous manuscript. We have thus showed that our simulated lake chlorophyll a concentrations generally fall within a range of in-situ measurements from globally distributed lakes and added discussion of the uncertainties associated with the algae dynamics.

Finally, we note several relevant additional modifications made in response to the other reviewer. Sensitivity analyses have been expanded substantially by perturbing all calibrated parameters. We have also added sensitivity simulations changing the fractions of prescribed TN and TP inputs assigned to different N and P species to the previous model component and input manipulations. All analyses have been limited to 2000 (instead of 2010), reflecting the limited availability of several components of required forcing data beyond 2000. For cross-watershed and global evaluations, annual results for the year 1990 are analyzed and presented along with ranges for the years 1990-2000. We have also added time series analyses of nutrient and solid loads from large U. S. rivers for the period ~1963-2000.

In this response, the line numbers correspond to those in the original manuscript before making any modifications. Table, figure, and section numbers have been changed due to additions/removals of tables, figures, and sections. Unless noted, the table, figure, and section numbers correspond to the modified ones. The modified tables and figures are placed at the end of this response.

Sincerely, Dr. Minjin Lee Associate Research Scholar Princeton University, NOAA/Geophysical Fluid Dynamics Laboratory (on behalf of all co-authors)

### Referee: 2

In this study, Lee et al. intend to develop a comprehensive freshwater model FANSY which can simulate the dynamics of algae, solid and nutrient dynamics in inland waters, and the fresh water model has been incorporated into the Land Model LM3. The topic is very important and should be attractive to a broad of readers. The paper is well organized and the methods and materials have been described adequately. This study involves tremendous of model development work. I appreciate the effects of the authors on developing such a model.

Nonetheless, I still have some concerns on the algorithms used in this study to simulate the transfer & transformation of sediment, N and P through the inland water systems. In specific:

1) To calculate the erosion-induced POC loss from soil to inland waters, why not calculate the POC loss rate based on the soil SOC concentration and the soil erosion rate, especially you are going to develop a process-based model? The empirical equation from Busen et al., 2005 might be too coarse to be used here for calculating the POC loss rate, and it cannot represent the effects of SOC dynamics on the riverine POC flux. In addition, many studies have proved the effectiveness of calculating POC loss rate based on the soil erosion rate and the SOC concentration, such as Tian et al., 2015 (Anthropogenic and climatic influences on carbon fluxes from eastern North America to the Atlantic Ocean: A process-based modeling study) and Zhang et al., 2022 (Estimating the lateral transfer of organic carbon through the European river network using a land surface model. Earth System Dynamics)

We agree with these points and have implemented your suggested change in approach. Please find the details in our combined response to your main comments below.

2) PON generally flows together with POC, as POC, PON and POP are all contained in the POM (particulate organic matter). Almost all particulate matter loss from land to rivers is caused by soil erosion, rather than the water drainage. Thus, it does not make sense to calculate PON loss based on the soil water drainage. A more reasonable solution is that, loss of all particulate matters (e.g. sediment, POC, POC, POP) from land to rivers should be calculated based on the soil erosion rate and the concentrations of these matters in soil; loss of all dissolved matters (DOC, DON, DOP) should be calculated based on soil water drainage (i.e. the leaching processes). Tian et al., 2015 & Zhang et al., 2022 can be good references.

Response to 1) & 2) We greatly appreciate the reviewer's incisive comments and informative references.

We agree that PON exports from land to rivers should be assumed to occur with eroded soil fluxes. We thus now simulate PON fluxes to rivers based on the simulated soil erosion fluxes

and litter/soil N concentrations, adapting an approach of previous studies that simulate POC fluxes based on soil erosion fluxes and soil C concentrations (Tian et al., 2015; Zhang et al., 2022).

We also now use the simulated PON fluxes to calculate inorganic soil fluxes from the total soil erosion fluxes, so that the eroded soil and PON inputs to rivers are self consistent. This now allows us to use inorganic suspended solids (ISS) in freshwaters as our prognostic variable, and derive total suspended solids (SS, i.e., the sum of ISS and POM) from the combination of freshwater ISS and PON. This requires an assumed ratio of PON to POM in eroded soils (details are provided below). Since the content of POM in eroded soils is generally small, our results proved insensitive to uncertainties in this conversion. The same ratio has been used to convert PON to POM in freshwaters for the purpose of calculating SS to compare with observations.

Since we do not model the freshwater C cycle yet, the corresponding POC fluxes are not explicitly simulated (though we note that an estimate is possible by using the simulated litter/soil C concentrations as done for PON fluxes). Similarly, since LM3 does not yet include terrestrial P dynamics, we have retained externally specified POP fluxes from Beusen et al (2015). We have extended the discussion of the need to pursue advances to provide a more comprehensive and consistent approach to modeling the coupled N, C, and P cycles across the terrestrial and freshwater continuum of LM3-FANSY.

In accordance with these changes, we now have removed equations (8)-(10), and (28), as well as the text describing our previous approach (lines 206-214, 289-302 in our initial submission) and modified Fig. 1.

And have replaced them with (line 162):

In LM3-FANSY, terrestrial soil erosion is controlled by land surface slope, rainfall, and leaf area index (LAI) based on Pelletier (2012), as described in Eq. (3). N fluxes from terrestrial litter and soils, in the form of PON, in Eq. (4) are simulated based on the simulated soil erosion fluxes and litter/soil N concentrations. This approach is consistent with that employed by several previous modeling studies (Tian et al., 2015; Zhang et al., 2022). The litter/soil concentrations for this purpose are estimated by using litter/soil contents and effective soil depths simulated by LM3 (Gerber et al., 2010).

Inorganic soil inputs to rivers are derived from the simulated soil erosion fluxes by subtracting the PON contribution, as described in Eq. (5). This requires an assumed ratio of POM:PON in eroded soils. Previous studies have shown a wide range of C content in tree biomass (~42-61%, Thomas and Martin, 2012) and of C:N ratios in litter and soils (~5-500, Gerber et al., 2010 and references in Gerber et al., 2010's Table S1). This implies that the POM:PON ratio in soil erosion fluxes can also vary significantly. We have found,

however, that predicted SS loads are insensitive to an order of magnitude variation in the ratio (i.e., 1.39 vs. 139, see Sect. 4.4), because organic contents in eroded soils are generally small. We thus used a POM:PON ratio of 13.9. The same ratio has been used to estimate the contribution of PON to SS in freshwaters, again noting that it is generally a small fraction of SS.

In accordance with these changes, we now have added/modified equations as follows:

$$\mathbf{E} = \mathbf{C}_1 \cdot \frac{\rho_b}{\rho_w} \cdot \mathbf{S}^{5/4} \cdot \mathbf{r} \cdot \mathbf{e}^{-\mathbf{L}} , \qquad (3)$$

$$\mathbf{E}^{\text{PON}} = \mathbf{E} \cdot \left(\frac{\mathbf{N}_{\text{FL}} + \mathbf{N}_{\text{SL}} + \mathbf{N}_{\text{SS}}}{\mathbf{h}_{\text{s}} \cdot \boldsymbol{\rho}_{\text{b}}}\right),\tag{4}$$

$$\mathbf{E}^{\mathrm{ISS}} = \mathbf{E} - \mathbf{r}_{\mathrm{DN,Ero}} \cdot \mathbf{E}^{\mathrm{PON}} \tag{5}$$

where E,  $E^{PON}$ , and  $E^{ISS}$  is terrestrial soil erosion flux (dry matter (D)kg m<sup>-2</sup> s<sup>-1</sup>), terrestrial PON flux (Nkg m<sup>-2</sup> s<sup>-1</sup>), and terrestrial inorganic soil erosion flux (Dkg m<sup>-2</sup> s<sup>-1</sup>) respectively, C<sub>1</sub> is a free parameter of soil erosion (unitless),  $\rho_b$  is soil bulk density (kgD m<sup>-3</sup>),  $\rho_w$  is water density(kg m<sup>-3</sup>), S is slope tan $\theta$ , with  $\theta$  as hillslope angle (unitless), r is rainfall (kg m<sup>-2</sup> s<sup>-1</sup>), L is LAI (unitless), N<sub>FL</sub>, N<sub>SL</sub>, and N<sub>SS</sub> is N content in fast litter, slow litter, and slow soil pool respectively (kgN m<sup>-2</sup>), h<sub>s</sub> is effective soil depth (m), and r<sub>DN,Ero</sub> is a POM-to-PON ratio in eroded fluxes (kgD kgN<sup>-1</sup>).

The equation for ISS which, as described above, is now a prognostic variable, is:

#### For a batch river and lake system, a mass balance for ISS and Sed is written as:

$$\frac{dISS}{dt} = \begin{cases} \frac{Sed}{dt} & R_{\#} < 1.2 \\ -\frac{w_{S}}{z} \left(\frac{1}{dt} + \frac{w_{S}}{z}\right)^{-1} \frac{ISS}{dt} & R_{\#} \ge 1.2 \end{cases},$$

$$\frac{dSed}{dt} = \begin{cases} -\frac{Sed}{dt} & R_{\#} < 1.2 \\ \frac{w_{S}}{z} \left(\frac{1}{dt} + \frac{w_{S}}{z}\right)^{-1} \frac{ISS}{dt} & R_{\#} \ge 1.2 \end{cases},$$
(8)
$$(9)$$

where ISS is inorganic suspended solid (kgD), Sed is benthic sediment inorganic solid (kgD), and z is river or lake depth (m).

The conversion of PON to POM in freshwaters for the purpose of calculating SS to compare with observations is as:

$$POM = r_{DN} \cdot PON,$$
(10)  

$$SS = ISS + POM,$$
(11)

## where POM, PON, and SS is particulate organic matter (kgD), particulate organic N (kgN), and suspended solid (kgD) respectively, and $r_{DN}$ is a POM-to-PON ratio in freshwaters (kgD kgN<sup>-1</sup>).

These modifications have changed model results. For solids, we have recalibrated the free parameter of terrestrial soil erosion ( $C_1$ ). The value was 0.015 and is now 0.012. After this relatively modest change, the modified SS results are very similar to the previous results. Correlations between the measurement-based and simulated SS yields, loads, and concentrations across the 64 rivers were 0.66, 0.77, and 0.67 respectively and now are 0.65, 0.76, and 0.67. The associated Fig. 2 and text in lines 448-449 have been modified as follows:

# Measurement-based and simulated annual SS estimates across 64 rivers are significantly correlated, with Pearson correlation coefficient, r values equal to 0.65 (0.57-0.65) for yields, 0.76 (0.71-76) for loads, and 0.67 (0.67-0.69) for concentrations for the year 1990 (range for the years 1990-2000) (Fig. 2, Table 4).

The total amount of global river SS loads to the coastal ocean was previously estimated as 9-11 Pg yr<sup>-1</sup> between 1982-2010, and now are 10 Pg yr<sup>-1</sup> for the year 1990 and 10-11 Pg yr<sup>-1</sup> for the years 1990-2000. The associated Table 5 and text in lines 456-459 have been modified as follows:

The total amount of global river SS loads to the coastal ocean estimated as 10 (10-11) Pg yr<sup>-1</sup> for the year 1990 (range for the years 1990-2000) by LM3-FANSY is at the lower bond of previous estimates (Table 5, Global NEWS estimates of 11-27 Pg yr<sup>-1</sup>, Beusen et al., 2005; Discharge Relief Temperature sediment delivery model (QRT) estimate of 12.6 Pg yr<sup>-1</sup>, Syvitski et al., 2005).

For N, the changes associated with the modified PON inputs to rivers are most manifest in river PON and TKN (i.e., the sum of PON, DON, and NH<sub>4</sub><sup>+</sup>) loads.

Correlations between the measurement-based and simulated TKN yields, loads, and concentrations across the 11 rivers were 0.57, 0.77, and 0.59 respectively and now are 0.49, 0.70, and 0.62. Global river PON loads to the coastal ocean have been reduced substantially from 14 (13-16) to 7 (7-8) for the year 1990 (range for the years 1990-2000). This has largely resulted from reduced PON inputs to rivers from 14 (14-16) to 4, which is likely at a lower bound, as both the simulated soil erosion fluxes and litter/soil N concentrations are at the lower bounds of published ranges. This result is further discussed later in this response and within the extended discussion. The associated Fig. 3, Tables 4-5, and text in lines 493-502 have been modified as follows:

Recent estimates of global river TN loads to the coastal ocean vary widely, ranging from about 36.5 to 48 TgN/yr (Table 5, Beusen et al., 2016; Boyer et al., 2006; Galloway et al., 2004; Green et al., 2004; Mayorga et al. 2010). Our global estimate 35 (34-38) TgN/yr for the year 1990 (range for the years 1990-2000) is at the lower bound of the published range. The simulated global river TN loads contain approximately equal contributions by DIN (the sum of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>, 14 (14-16) TgN yr<sup>-1</sup>, 41% of TN) and DON (13 (13-14) TgN yr<sup>-1</sup>, 39% of TN), with a lesser contribution by PON (7 (7-8) TgN yr<sup>-1</sup>, 20% of TN). The estimates of global river DIN loads are at the lower end of recent estimates, which range from 14.5 to 18.9 TgN yr<sup>-1</sup> (Mayorga et al. 2010; Green et al., 2004; Smith et al., 2003). This may be partly due to an overestimate of freshwater denitrification and/or algae-mediated transformations to organic forms. In contrast, our global river DON load estimate is slightly higher than a previous estimate 10 TgN yr<sup>-1</sup> (Harrison et al., 2005). Our global river PON load estimate is considerably lower than a previous estimate, 13.5 TgN yr<sup>-1</sup> (Mayorga et al. 2010). See Sect. 4.5 for further discussion.

The discussion of the need to provide a more comprehensive and consistent approach of modeling the coupled N, C, and P cycles in the terrestrial and freshwater continuum of LM3-FANSY was in lines 670-672 (which has been moved under a newly added subsection Sect. 4.5 in the modified manuscript) and reads as follows:

In addition, there is a need to pursue advances to provide a more comprehensive and consistent approach of modeling the coupled N, C, and P cycles across the terrestrial and freshwater continuum of LM3-FANSY. Expansion of LM3 to include terrestrial P dynamics will be targeted to improve estimates of litter/soil P storage and fluxes to streams and rivers, generating mechanistically consistent estimates of N:P ratios of nutrient loads reaching coastal systems. Priority enhancements will also include integration of freshwater C and alkalinity dynamics with the current solid, algae, and nutrient dynamics of FANSY to simulate the impacts of river inputs on coastal C budgets and acidification.

The following references are newly added.

Tian, H., Yang, Q., Najjar, R. G., Ren, W., Friedrichs, M. A. M., Hopkinson, C. S., and Pan, S.: Anthropogenic and climatic influences on carbon fluxes from eastern North America to the Atlantic Ocean: A process-based modeling study, J. Geophys. Res. Biogeosci., 120, 757–772, 2015.

Zhang, H., Lauerwald, R., Regnier, P., Ciais, P., Van Oost, K., Naipal, V., Guenet, B., and Yuan, W.: Estimating the lateral transfer of organic carbon through the European river network using a land surface model, Earth Syst. Dynam., 13, 1119–1144, https://doi.org/10.5194/esd-13-1119-2022, 2022. 3) To my knowledge, riverine DON mostly originated from soil during the leaching processes. Decomposition of PON and algae mortality only contribute a small part of the total DON. However, the FANSY model in this study assumed all DON is originated from the decomposition of PON and algae mortality. This assumption does not make sense. I would suggest the authors to add the fluxes of DOC, DON and DOP from land to river during the leaching processes. Many studies have revealed that the fluxes of dissolved matters caused by leaching is much larger than the loss of particulate matters caused by soil erosion (Regnier et al., 2022; Nature, The land-to-ocean loops of the global carbon cycle)

We apologize for the misunderstanding: LM3 simulates soil  $NO_3^-$ ,  $NH_4^+$ , and DON leaching. We have clarified input fluxes from terrestrial system and the atmosphere to rivers in Fig. 1 and on lines 114-116. Consistent with the reviewer's view, river DON loads to the coastal ocean estimated as 13 TgN yr<sup>-1</sup> are mostly originated from soil DON leaching (49 TgN yr-1) and the sum of soil  $NO_3^-$ ,  $NH_4^+$ , and DON leaching (74 TgN yr-1) is much greater than the PON flux to rivers (4 TgN yr-1).

As noted in the previous response, since we do not model the freshwater C cycle yet, we did not explicitly simulate soil DOC leaching, and POP leaching is handled through input estimates from Beusen et al (2015). This simplification is largely because the land model, LM3, does not yet include a P component.

4) I am surprised that the model does not consider the emission of N2O from inland waters. Both observation and simulation indicated that a larger part of the riverine N is loss to air in the form of N2O (Yao et al., Nature Climate Change, Increased global nitrous oxide emissions from streams and rivers in the Anthropocene)

We have included denitrification emissions from freshwaters, but have not included processes of explicitly separating N<sub>2</sub>O and N<sub>2</sub> emissions from the freshwater denitrification. For this initial model development, simulating the total amount of N removal to the atmosphere via the freshwater denitrification has been sufficient for the purpose of closing the freshwater N budget and estimating the model's capacity to capture globally distributed N and P loads to the coastal ocean. We agree that freshwater N<sub>2</sub>O emission has been recognized as an increasingly important greenhouse gas source, and thus N<sub>2</sub>O modeling should be subject to future work.

We have added associated text in line 670 (which has been moved under a newly added subsection Sect. 4.5 in the modified manuscript) as follows:

There is also significant room for further model development and improvement. The current version of LM3-FANSY simulates denitrification emissions from freshwaters, but does not include processes that explicitly separate N<sub>2</sub>O and N<sub>2</sub> emissions from the freshwater denitrification. As freshwater N<sub>2</sub>O emissions have been recognized as an increasingly important greenhouse gas source (Yao et al., 2020), it will be important to differentiate N<sub>2</sub> and N<sub>2</sub>O emission processes in future work.

The following reference is newly added.

Yao, Y., Tian, H., Shi, H. et al.: Increased global nitrous oxide emissions from streams and rivers in the Anthropocene, Nat. Clim. Chang., 10, 138–142, https://doi.org/10.1038/s41558-019-0665-8, 2020.

5) Many of the forcing data used in this study were obtained from Beusen et at., 2015. It is better to provide more information about these data. Foe example, the spatial and temporal resolutions of these data, and the method used by Beusen et al., 2015 to produce these data.

We have extended the description of data from Beusen et al (2015) in lines 402-403 as follows:

Solid and nutrient inputs from terrestrial systems and from the atmosphere to rivers are either simulated by LM3-FANSY or provided by Beusen et al (2015) (Table 3). For N, all inputs were simulated by LM3-FANSY except aquaculture, wastewater, and atmospheric deposition, which were provided by Beusen et al (2015). For P, which is not currently included in LM3, all inputs were provided by Beusen et al (2015).

Beusen et al (2015) provided five-year interval data for the period 1900-2000 at 0.5 degree resolution. The data were regridded to our 1 degree resolution by summing up the values given in kg yr<sup>-1</sup> and linearly interpolated across the five-year intervals. Beusen et al (2015)'s wastewater N and P inputs were from Morée et al (2013)'s urban waste N and P discharge estimates to surface waters. Beusen et al (2015) calculated aquaculture N and P inputs using Bouwman et al (2013)'s finfish and Bouwman et al (2011)'s shellfish data. For atmospheric N deposition inputs, Beusen et al (2015)'s input for the year 2000 was from Dentener et al (2006)'s ensemble of reactive-transport models and those for the years before 2000 were made by scaling the deposition with Bouwman et al (2013)'s ammonia emissions. Beusen et al (2015)'s surface runoff P inputs include those leached from soil P budgets (i.e., the sum of fertilizer and animal manure minus crop and grass withdrawal) and those driven by soil erosion estimates based on Cerdan et al (2010). Beusen et al (2015)'s P inputs of litter from floodplains were estimated as 50% of total NPP with a C:P

ratio of 1200. Beusen et al (2015)'s weathering P inputs were computed based on Hartmann et al (2014)'s chemical weathering P release estimates.

The following references are newly added.

Bouwman, A. F., Beusen, A. H. W., Overbeek, C. C., Bureau, D. P., Pawlowski, M., and Glibert, P. M.: Hindcasts and future projections of global inland and coastal nitrogen and phosphorus loads due to finfish aquaculture, Rev. Fish Sci., 21, 112–156, doi:10.1080/10641262.2013.790340, 2013.

Bouwman, A. F., Klein Goldewijk, K., Van der Hoek, K. W., Beusen, A. H. W., Van Vuuren, D. P., Willems, W. J., Rufino, M. C., and Stehfest, E.: Exploring global changes in nitrogen and phosphorus cycles in agriculture induced by livestock production over the 1900–2050 period, P. Natl. Acad. Sci. USA, 110, 20882–20887, doi:10.1073/pnas.1012878108, 2013.

Bouwman, A. F., Pawłowski, M., Liu, C., Beusen, A. H. W., Shumway, S. E., Glibert, P. M., and Overbeek, C. C.: Global hindcasts and future projections of coastal nitrogen and phosphorus loads due to shellfish and seaweed aquaculture, Rev. Fish Sci., 19, 331–357, doi:10.1080/10641262.2011.603849, 2011.

Cerdan, O., Govers, G., Le Bissonnais, Y., Van Oost, K., Poesen, J., et al.: Rates and spatial variations of soil erosion in Europe: A study based on erosion plot data, Geomorphology, 122, 167–177, doi:10.1016/j.geomorph.2010.06.011, 2010.

Dentener, F., Stevenson, D., Ellingsen, K., Noije, T. v., Schultz, M., et al.: The global atmospheric environment for the next generation, Environ. Sci. Technol., 40, 3586–3594, 2006.

Hartmann, J., Moosdorf, N., Lauerwald, R., Hinderer, M., andWest, A. J.: Global chemical weathering and associated P-release – The role of lithology, temperature and soil properties, Chem. Geol., 363, 145–163, doi:10.1016/j.chemgeo.2013.10.025, 2014.

Morée, A. L., Beusen, A. H. W., Bouwman, A. F., and Willems, W. J.: Exploring global nitrogen and phosphorus flows in urban wastes during the twentieth century, Global Biogeochem. Cy., 27, 1–11, doi:10.1002/gbc.20072, 2013.

6) Discussion on the uncertainties of this study is weak. Consider the simplification and ignorance of many processes that are closely related to the transfers of sediment, C, N & P. There should be more discussion on the potential uncertainties in the simulated results. For example, the uncertainties in simulated soil erosion rate, POC loss rate and the effects of dams and reservoirs.

We agree that the discussion and analyses of uncertainties generally, and in particular those of modeling soil erosion and associated fluxes to rivers and the coastal ocean were not sufficient in the previous manuscript.

In the modified manuscript, we have ensured that the uncertainties and simplifications are highlighted through the Methods and Results, and we have extended the discussion of the uncertainties, including the specific issues the reviewer has raised, by adding a new subsection 4.5 as follows:

### 4.5 Discussion on uncertainties and future work

The inputs and transport of solids and nutrients through the terrestrial-freshwater system, and transformations within it are governed by complex and interlinked physical, chemical, and biological processes. The understanding of these processes varies greatly, as does the degree of their inevitable simplifications within LM3-FANSY. We have highlighted the numerous uncertainties and simplifications in the model description and result presentation. Here, we discuss the prominent uncertainties that will be prioritized in future work.

There are several significant uncertainties in modeling the soil erosion and associated fluxes of solids and PON to rivers and the coastal ocean. Our global river PON load estimate (7, 7-8 Tg yr<sup>-1</sup>) is lower than that of Global NEWS 2 (13.5 Tg yr<sup>-1</sup>, Table 5), but confidences in both estimates are low, without explicit evaluations due to relatively limited direct measurements of PON. Our estimate, however, may be relatively low, when considering that both simulated global river SS loads (10, 10-11 Pg yr<sup>-1</sup>) and global litter/soil N storage (86 Pg) in LM3-FANSY are at the lower bounds of previous estimates (11-27 Pg yr<sup>-1</sup> for SS loads, Table 5 and 95 (70-820) Pg for N storage, Post et al., 1985). While river TKN loads, which include PON, agree fairly well with the measurement-based estimates across various rivers (Fig. 3), it seems probable that our estimate is on the low end. Several factors may have contributed to this. The current relatively coarse resolution globally implemented herein inevitably "glosses over" areas of peak soil erosion. The single vertical layer formulation of LM3 omits any potential interactions between the vertical distribution of soil erosion and that of litter and soil N storage. An implementation of LM3-FANSY at higher resolution capturing the larger number of rivers will allow an expansive evaluation against the larger number of observations, and facilitate a better assessment of these uncertainties.

The challenges of modeling particulates continue once they have entered the freshwater system. The Rouse number-dependent transport criterion from Pelletier (2012) was adapted to simulate deposition/resuspension fluxes between suspended matters (i.e., ISS, PON, POP and PIP) and benthic sediments (i.e., Sed, SedN, and SedP). The criterion was designed to primarily simulate suspended loads, typically accounting for > 80% of total (i.e., suspended and bed) loads from most large (> ~100 km2) river basins (Pelletier, 2012; Turowski et al., 2010), without explicitly modeling benthic sediments. We acknowledge that our simplified benthic sediment component resulted from adapting the Pelletier's approach drives uncertainties in modeling suspended matters and benthic sediments, including the important biogeochemical transformations, such as denitrification, that occur within the sediments. An implementation of more sophisticated benthic sediment dynamics and bed load transport processes is thus subject to critical future work.

Uncertainties associated with sediment dynamics and bed load transport are compounded by the relatively simple representation of lakes, and the exclusion of anthropogenic hydraulic controls like damming, irrigation, and diversion that affect many rivers. For model evaluation, if available, we used the natural water discharges of Meybeck and Ragu (2012) when calculating loads and yields from their multi-year average concentrations (see Sect. 3.3). Large dams or reservoirs, however, have been shown to impound solids and nutrients to substantially decrease their loadings to rivers (Vorosmarty et al., 2003). Thus, despite the relatively low global river SS loads in this first implementation of LM3-FANSY, the lack of such sediment trapping may have induced overestimations of solid and nutrient loads from river basins including large dams or reservoirs. As a representative example, the Colorado River Basin is known for nearly complete trapping of solids due to large reservoir construction and flow diversion (Vorosmarty et al., 2003). LM3-FANSY does not capture such an extreme trapping. As a result, the Colorado River SS load simulated by LM3-FANSY (99,235 kt yr<sup>-1</sup>) is more consistent with the corresponding load calculated by using the "natural" water discharge of Meybeck and Ragu (2012) (120,010 kt yr<sup>-1</sup>) than with the load calculated by using the "actual" water discharge (649 kt yr<sup>-1</sup>). Although use of the actual water discharges is found to not significantly alter the cross-watershed evaluations (Fig. SI6), such anthropogenic hydraulic controls are expected to further increase in the future (Seitzinger et al., 2010). It will be thus important to consider the effects of such controls in future work.

There is also significant room for further model development and improvement. An improved representation of lakes (e.g., vertical layering) is necessary to better resolve algal processes and associated transformations between inorganic and organic phases.

Modeling large lakes with the ocean component of GFDL's Earth System Model (Adcroft et al., 2019) is one of our priority developments, particularly given the importance of algae as a control on the relative proportions of inorganic vs. organic nutrients in freshwaters. An initial configuration for the U. S. Great Lakes is currently under development.

There is also a need to pursue advances to provide a more comprehensive and consistent approach of modeling the coupled N, C, and P cycles across the terrestrial and freshwater continuum of LM3-FANSY. Expansion of LM3 to include terrestrial P dynamics will be targeted to improve estimates of litter/soil P storage and fluxes to streams and rivers, generating mechanistically consistent estimates of N:P ratios of nutrient loads reaching coastal systems. Priority enhancements will also include integration of freshwater C and alkalinity dynamics with the current solid, algae, and nutrient dynamics of FANSY to simulate the impacts of river inputs on coastal C budgets and acidification.

The current version of LM3-FANSY simulates denitrification emissions from freshwaters, but does not include processes that explicitly separate N<sub>2</sub>O and N<sub>2</sub> emissions from the freshwater denitrification. As freshwater N<sub>2</sub>O emissions have been recognized as an increasingly important greenhouse gas source (Yao et al., 2020), it will be important to differentiate N<sub>2</sub> and N<sub>2</sub>O emission processes in future work.

Although LM3-FANSY is capable of producing river solids and nutrients, in various forms and units, some disagreements between the modeled and measurement-based estimates remain. Many observational studies have noted the uncertainties associated with measurement methods, location, and frequency that likely contribute to these disagreements. Additional time varying constraints are also needed to build additional confidence in projected changes. Finally, all of these model improvement efforts will be greatly facilitated by implementing LM3-FANSY at higher resolution capturing the larger number of rivers and by extensive river measurements across the world, with a better assessment of uncertainties.

The following references are newly added.

Adcroft, A., Anderson, W., Balaji, V., Blanton, C., Bushuk, M., Dufour, C. O., et al.: The GFDL global ocean and sea ice model OM4.0: Model description and simulation features, J. Adv. Model. Earth Syst., 11, 3167–3211, <u>https://doi.org/10.1029/2019MS001726</u>, 2019.

Post, W. M., Pastor, J., Zinke, P. J., and Stangenberger, A. G.: Global patterns of soil nitrogen storage, Nature, 317, 613–616, 1985.

Turowski, J. M., Rickenmann, D., and Dadson, S. J.: The partitioning of the total sediment load of a river into suspended load and bedload: A review of empirical data, Sedimentology, 57, 1126–1146, doi:10.1111/j.1365-3091.2009.01140.x, 2010.

7) Compared to existing models, a highlight point of the FANSY model in this study is that it explicitly simulated the biogeochemical cycles related to the algae. However, there is no evaluation of the simulated algae processes, and it is hard for the readers to determine whether the scheme used in the FANSY model is effective or reliable on simulating algae dynamics. Is it possible for the authors to collect some observation data to validate the simulated algae biomass, growth rate or mortality rate in this study.

A detailed comparison of LM3-FANSY results against observed chlorophyll a is difficult, because there are no compiled estimates of chlorophyll a in rivers. While Sayers et al (2015) provides a compilation of lake chlorophyll a estimates, many lakes are small and those that are large can have highly heterogenous chlorophyll a distributions. This poses a challenge, given the relatively simple nature of lake biogeochemistry in LM3-FANSY (i.e., vertically unresolved mixed reactors, Lee et al., 2019) and the current coarse global resolution. However, in the Results, we now provide a map of the simulated lake chlorophyll a concentrations (Fig. SI3) and confirm that the range of simulated values are comparable to that in Sayers et al (2015). We have also expanded our sensitivity analysis of algae dynamics to better elucidate the controls that algae exert on the ratios of inorganic to organic nutrients. Finally, we have identified an improvement of the lake representation as a future development priority (please refer to our response to your previous comment 6).

The chlorophyll a result has been added in Sect. 4.1 Model performance analysis, as follows:

Despite the relatively simple nature of lake biogeochemistry in LM3-FANSY (i.e., vertically unresolved mixed reactors, Lee et al., 2019), the model creates a reasonable range of chlorophyll a concentrations (Fig. SI3) that generally fall within a range of in-situ estimates from globally distributed lakes (Sayers et al., 2015). The in-situ estimates in the compilation of Sayers et al. (2015) range from ~0 to ~100 mg m<sup>-3</sup>, mostly falling between 5 and 50 mg m<sup>-3</sup> (Fig. 5 of Sayers et al. (2015), available at https://www.tandfonline.com/doi/full/10.1080/01431161.2015.1029099).

We note that we have slightly decreased algal mortality rate constant from 1.0 10-5 to 0.8 10-5 kgN<sup>-1/3</sup>s<sup>-1</sup>. The text describing the expanded sensitivity tests, highlighting the algae controls on partitioning of inorganic and organic nutrients (line 651) is as follows:

Algae dynamics play a significant role in determining the relative composition of inorganic vs. organic nutrients in freshwaters. Decreasing the algal mortality rate constant by half enhances algal uptake, decreasing DIN (the sum of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>) and IP (the sum of PO4<sup>3-</sup> and PIP) by -11% and -33% respectively, while it increases ON (the sum of DON and PON) and OP (the sum of DOP and POP) by 24% and 33% respectively. Similarly, increasing the maximum photosynthesis rate or chlorophyll a-specific initial slope of the photosynthesis-light curve by twice enhances algal uptake, decreasing DIN by -6 or -7% and IP by -21% or -25% respectively, while it increases ON by 18% or 21% and OP by 23% and 28% respectively. The opposite holds for the parameter changes that reduce algal uptake. An analysis of the model sensitivity simulations, wherein the dynamic contributors to light extinction (i.e., ISS, POM, and CHL in Eq. (23)) were removed, further suggests that the proper light limitation of algal growth is also important for skillful estimates of freshwater inorganic vs. organic nutrients. Removing the dynamic light shading component leads to a ~26 % and ~35% overestimation of ON and OP loads and an underestimation of DIN and IP loads by ~10 % and ~32% respectively. Inorganic nutrient levels are suppressed by invigorated algal populations and more nutrients end up in organic forms. Algal controls thus offer an effective means of calibrating the mix of inorganic and organic constituents.

The following reference is newly added.

Sayers, M. J., Grimm, A. G., Shuchman, R. A., Deines, A. M., Bunnell, D. B. et al.: A new method to generate a high-resolution global distribution map of lake chlorophyll, Int. J. Remote Sens., 36, 1942-1964, DOI: <u>10.1080/01431161.2015.1029099</u>, 2015.

### Minor comments:

### In Eq. 3, how is the slope extracted? Is the slope extracted from DEM at 1-degree resolution?

The slope was created by using the Global Multi-resolution Terrain Elevation Data 2010 (GMTED 2010) at 1/10-degree resolution, but we had to regrid it to 1-degree resolution by averaging out to match the model resolution.

Danielson, J. J. and Gesch, D. B.: Global multi-resolution terrain elevation data 2010 (GMTED2010): U.S. Geological Survey Open-File Report 2011–1073, https://pubs.usgs.gov/of/2011/1073/pdf/of2011-1073.pdf, 2011.

### In Eq. 4, how the depth of river and lake is calculated? Do you have a map of area and lake as the forcing data of your model? If yes, is the area of river or lake fixed?.

Shallow lake depths were fixed as 2 m. For large lakes, the depths were calculated by dividing lake volumes by lake surface areas based on data from van der Leeden et al. (1990). Maximum depths calculated in this way were limited to 50 m. Lake areas and lake area fractions of grid cells were prescribed, defined on the basis of the U. S. Geological Survey Global Land Cover Characteristics Database "IGBP Water Bodies" field (Please see the maps attached in this response).

River lengths were prescribed, but river depths, widths, and velocities were simulated at every time step (30 minutes) based on simulated river flows, yielded from hydraulic geometric relations of Leopold and Maddock (1953).

A detailed description of river and lake hydrology and hydrography can be found in Milly et al. (2014).

Milly, P. C. D., Malyshev, S., Shevliakova, E., Dunne, K. A., Findell, K. L., Gleeson, T., Liang, Z., Phillipps, P., Stouffer, R. J., and Swenson, S.: An enhanced model of land water and energy for global hydrologic and earth-system studies, J. 845 Hydrometeorol., 15, 1739–1761, 2014.



Figure 1: LM3-FANCY structure with arrows depicting fluxes of constituents of algae, nutrients, and solids in rivers and lakes. The constituents are listed in Table 1.



Figure 2: Pearson correlation coefficients (r) and p values (p) between the measurementbased vs. simulated SS yields, loads, and concentrations across 64 rivers for the year 1990.



**Observed**, Log-scale

Figure 3: Pearson correlation coefficients (r) and p values (p) between the measurementbased vs. simulated NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, DON, and TKN yields, loads, and concentrations across 50, 36, 18, and 11 rivers for the year 1990.



**Observed**, Log-scale

Figure 4: Pearson correlation coefficients (r) and p values (p) between the measurementbased vs. simulated PO4<sup>3-</sup>, DOP, and TP yields, loads, and concentrations across 46, 9, and 5 rivers for the year 1990.

|                     |                  | SS                   | NO <sub>3</sub> -       | $NH_4^+$                | DON               | TKN                 | PO4 <sup>3-</sup>       | DOP               | TP                |
|---------------------|------------------|----------------------|-------------------------|-------------------------|-------------------|---------------------|-------------------------|-------------------|-------------------|
| r                   |                  | .76<br>(.71, .76)    | .79<br>(.75, .81)       | .66<br>(.65, .74)       | .85<br>(.85, .93) | .70<br>(.65, .74)   | .70<br>(.66, .74)       | .93<br>(.93, .95) | .98<br>(.95, .98) |
| NSE                 |                  | .55<br>(.48, .55)    | .42<br>(.33, .49)       | .37<br>(.34, .49)       | .66<br>(.62, .77) | .00 (.00, .22)      | .48<br>(.42, .53)       | .85<br>(.85, .88) | .82<br>(.81, .86) |
| Prediction<br>error | Min              | -97<br>(-98, -97)    | -86<br>(-92, -79)       | -76<br>(-90, -76)       | -75<br>(-76, -68) | -56<br>(-60, -24)   | -98<br>(-99, -86)       | -51<br>(-55, -47) | -64<br>(-66, -61) |
|                     | 25 <sup>th</sup> | -35<br>(-53, -35)    | 1 (-12, 8)              | -48<br>(-49, -34)       | -43<br>(-48, -7)  | 31<br>(-8, 31)      | -59<br>(-62, -51)       | -39<br>(-50, -38) | -62<br>(-65, -60) |
|                     | Med              | 39<br>(18, 50)       | 146<br>(78, 203)        | 18<br>(-1, 45)          | 35<br>(2,75)      | 60<br>(33, 68)      | -22<br>(-29, -5)        | -3<br>(-17, 18)   | -10<br>(-16, 0)   |
|                     | 75 <sup>th</sup> | 260<br>(227, 339)    | 475<br>(396, 693)       | 540<br>(300, 541)       | 243<br>(140, 293) | 287<br>(259, 396)   | 171<br>(137, 234)       | 155<br>(114, 155) | 78<br>(26, 78)    |
|                     | Max              | 3219<br>(2324, 4238) | 1796<br>(1346,<br>3256) | 2946<br>(1099,<br>4158) | 610<br>(358, 733) | 1040<br>(911, 1374) | 1956<br>(1956,<br>2955) | 365<br>(274, 449) | 182<br>(103, 208) |
|                     | IQR              | 296<br>(273, 384)    | 474<br>(405, 705)       | 589<br>(349, 589)       | 286<br>(176, 317) | 256<br>(248, 383)   | 230<br>(198, 293)       | 194<br>(156, 194) | 141<br>(88, 141)  |

Table 4: Pearson correlation coefficients (r) and Nash–Sutcliffe model efficiency coefficient (NSE) between the measurement-based vs. simulated loads across world major rivers for the year 1990 (range for the years 1990-2000 in parenthesis). The prediction error is computed as the difference between the simulated and measurement-based estimates of loads expressed as a percentage of the measurement-based load.

|  | SS  | TN  | DIN   | DON  | PON   | TP  | DIP   | DOP   | PP  |
|--|---|---|---|--|---|---|---|---|---|
| LM3-FANSY  | 10<br>(10, 11)                              | 35<br>(34, 38)                                | 14<br>(14, 16)                                | 13<br>(13, 14)                                   | 7<br>(7, 8)                                   | 7<br>(7, 8)                                   | 2   | 1   | 5   |
| Global NEWS 1<br>(year 1995)<br>Global NEWS 2<br>(year 2000) | 19<br>(11, 27)<br>Beusen<br>et al<br>(2005) | 44.9<br>NEWS 2,<br>Mayorga<br>et al<br>(2010) | 18.9<br>NEWS 2,<br>Mayorga<br>et al<br>(2010) | 10<br>NEWS-<br>DON,<br>Harrison et<br>al. (2005) | 13.5<br>NEWS 2,<br>Mayorga<br>et al<br>(2010) | 9.04<br>NEWS 2,<br>Mayorga<br>et al<br>(2010) | 1.45<br>NEWS-<br>DIP-HD,<br>Harrison<br>et al<br>(2010) | 0.6<br>NEWS-<br>DOP,<br>Harrison<br>et al<br>(2005) | 6.56<br>NEWS 2,<br>Mayorga<br>et al<br>(2010) |
| QRT;<br>(years 1960-1995)                                    | 12.6<br>Syvitski<br>et al<br>(2005)         |   |   |  |   |   |   |   |   |
| IMAGE-GNM<br>(year 2000)                                     |   | 36.5<br>Beusen et<br>al (2016)                |   |  |   | 4<br>Beusen et<br>al (2016)                   |   |   |   |
| Boyer et al. (2006)<br>(mid-1990s)                           |   | 48  |   |  |   |   |   |   |   |
| Galloway et al.<br>(2004)<br>(early-1990s)                   |   | 47.8  |   |  |   |   |   |   |   |
| Green et al., 2004<br>(mid-1990s)                            |   | 40  | 14.5  |  |   |   |   |   |   |
| Smith et al., 2003<br>(1990s)                                |   |   | 18.9  |  |   |   | 2.3   |   |   |

Table 5: LM3-FANSY and published estimates of global river loads to the coastal ocean in Pg yr<sup>-1</sup> for SS and Tg yr<sup>-1</sup> for nutrients. LM3-FANSY results are for the year 1990 (ranges for the years 1990-2000).



Figure SI3. Chlorophyll a concentrations (mg m<sup>-3</sup>) in lakes for the year 1990.



Figure SI6: Pearson correlation coefficients (r) and p values (p) between the measurementbased vs. simulated yields, loads, and concentrations across the world major rivers for the year 1990. Here the actual (instead of natural) water discharges of Meybeck and Ragu (2012) were used when calculating loads and yields from their multi-year average concentrations.



Lake areas and lake area fractions of grid cells (in response to the last minor comment).

0 0 1 0

0 aa 1 0.000

180°



0° LONGITUDE

60°E

120°E

40**•**S

180°

120°W

60°W