Response to reviewer 1

Title: Global riverine N and P transport to ocean increased during the twentieth century despite increased retention along the aquatic continuum

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We are very grateful to the two reviewers for their constructive feedback. The suggestions definitely led to significant improvements of our text, particularly where the reviewers found sections that were not clear enough, or not quantitative (the sensitivity analysis, reviewer 1).

The concerns have been addressed below and in the revised manuscript. Below are the reviewer comments, **our response in bold**, and the new text that will be included in the revision of our paper, which is found between "".

REVIEWER 1

Reviewer 1. GENERAL COMMENTS

The study by Beusen et al. explores nutrient retention (N and P) and export to the ocean by the world's freshwater systems, giving estimates of the total riverine transport and assessing the main contributing factors to these global fluxes. The authors analyze long-term changes using a worldwide, spatially explicit model, and discuss the shift in nutrient sources and in-stream dynamics in relation with the changes in human activities throughout the 20th century. I believe it is a valuable paper, and it represents a relevant scientific contribution within the scope of Biogeosciences. I therefore recommend its publication with a few minor corrections.

Positive aspects of the manuscript are:

- The model analyzes nutrient dynamics taking into account hydrological changes, land-use factors, wastewater discharge, and in-stream processes. This allows for a good critical analysis of the impact of major human transformations over the past century.

- As stressed by the authors, the model considers the in-stream dynamics of P, something that is seldom, if ever, considered in similar modeling studies.

- Estimates on global N and P retention and export to the ocean are provided for the whole 20th century. Although modeled data is approximate and it should be further tested in many different freshwater systems, these integrated values are very useful for discussing changes in global biogeochemical cycles and in budget calculations.

There are a few issues that may need further discussion, or clarification:

1. - I understand that the model is described in full length in Beusen et al. (2015), and the information provided in the supplementary material is fairly complete, so there is no point repeating all such

information in the manuscript. Some particulars, however, are required to understand how the model works and what computation is behind the results. In this regard, some brief details on how the two models (IMAGE-GNM and PCR-GLOBWB) are coupled would be welcome.

2. - Given that the model uses a 0.5 by 0.5 grid, I agree with the authors that there is no point including small rivers in the analyses. They set the threshold in rivers < 10000 km2 (page 20129, line 3), but I wonder whether this may be a huge constraint in certain areas of the world, such as the Mediterranean, where the hydrological system is composed of many small streams, in most cases much smaller than the 10 000 km2 here established. Could the authors please discuss what might be the effect of neglecting such small rivers?

Response: We apologize, this is a misunderstanding. Rivers with basin areas <10,000 km^2 are not excluded in the model, but are excluded from the validation with measurement data. We modified the text to make this clear.

3. - In my opinion, a more in-depth comparison between some of the results of the model and actual data provided in other studies would be desirable. References could be given, for instance, to support the observed decrease in the N:P soil molar ratio (page 20129), the global increase in point sources (page 20131), the large nutrient retention in the Mediterranean and the Black Sea with regard to other areas (pages 20132-33), etc. This would actually serve as an additional model validation.

Response: The remark about the decrease of soil N:P ratio was incorrect and was therefore removed. We changed the text which now states: "The N:P ratio in fertilizers has been increasing since the 1970s (FAO, 2015). However, this change has been compensated by the expansion of livestock production, which produces high-P manure (Bouwman et al., 2013c)." We added two references supporting the wastewater N and P increase (Van Drecht et al., 2009; Moree et al., 2013). Unfortunately we have not found references that support our estimates of retention of N and P in the Mediterranean region. The Van Drecht et al. reference was added to the literature list.

4. I understand that long time series of riverine nutrient concentrations are not easy to find, yet it seems to me that validating world simulations with just three rivers, all of them located in the northern hemisphere and with similar climatological conditions (Fig. 2) is too limited. Have the authors tested some other rivers? If so, could them include them in the supplementary materials?

Response: In our GMD paper we compared model data with monitoring data for 125 European stations from EEA, 11 stations in the Mississippi and the rivers Meuse and Rhine. In this paper we show three examples of rivers not shown in the GMD paper. To make this clear we changed the text to: "Beusen et al. (2015) compared model results with the discharge-weighed annual mean calculated from long-term time series (from 1970 onwards to most recent years, depending on the station) of observed concentrations and discharge *for 125 European rivers*, and for the river Mississippi (11 stations), and the rivers Rhine and Meuse. *In this paper we show details of the model predictions and compare those with long-term time series for stations in the Danube in Hungary, Missouri in the USA and Ångermanälven in Sweden (Figure 2)."* Since the Missouri comparison is in the Supporting Information of the GMD paper, we added this to the figure caption: "Figure 2a is modified from Beusen et al., 2015)."

SPECIFIC COMMENTS and TECHNICAL CORRECTIONS

5. Abstract, Page 20124, Lines 8, 9, 14: Please correct the P units (5 to 9 Tg P yr-1; 3 to 5 Tg P yr-1; 2 to 4 Tg P yr-1).

Response: We thank the reviewer for this comment, we made the correction.

6. Page 20127, Lines 12-17: Is ammonia volatilization considered among the N outputs? Is it assumed to be globally offset?

Response: Ammonia volatilization is a loss term in the soil budget. Hence, both deposition and emissions are accounted for. The text now reads: "Nutrient outputs account for withdrawal by agricultural crops in harvested parts and by grazing or mowing of grass and ammonia volatilization."

7. Page 20127, Lines 22-23: the "memory" or retention time of groundwater, does it change much between world areas? (Groundwater represents a big share of the nutrient sources in Fig. 3, so I wonder whether there are important geographical differences).

Response: This is an important issue. The following sentence was added: "Cumulative N storage in deep groundwater between 1900 and 2000 amounted to around 376 Tg (Bouwman et al., 2013a). The retardation due to this cumulative reservoir varies considerably depending on the history of fertilizer use and manure management, as well as the geohydrological situation and climate (Van Drecht et al., 2003). "

8. Page 20127, Line 26: Wouldn't it be better to validate the model sensitivity for the year 1975 instead of the 1950, taking into account that it was mostly in the 70s when the largest nutrient increases, mainly from agricultural sources, took place (the steepest increasing slopes in Figs. 3-4-5 occur around the 70s)?

Response: We agree that the sensitivity for 1975 will be different from that in 1950. In 1975, fertilizer use in the industrialized countries and the former Soviet Union was at its peak, and since then it has been decreasing, whereas it has been increasing in China and India. However, we believe that comparing the situation right after World War II (1950), which is prior to the exponential increase of the world population, food, energy production and fertilizer use, with that in the year 2000 with a world population of over 6 billion inhabitants and fertilizer use of >80 million tonnes gives a clear picture of the model behavior for these two situations. The model sensitivity for the year 1975 would not be very different from 2000.

9. Page 20134: The authors compare the influence of the different factors for the N and P budgets over time. The discussion is somewhat vague, though: they say that the influence of this or that factor became important after a certain year, but how important? could they be more specific and broadly quantify such importance (e.g. did it represent over 30% of the total sources? half of the inputs?)?

Response: the discussion was vague. Tables SI4 and SI5 are explicit about the importance of model parameters, expressed by the SRC. We therefore made the text more explicit by adding the way parameters varied (mostly + or – 10%), and by adding the SRC values, which provide the impact on the model output (delivery, retention, export). To illustrate this we have added the following text to the manuscript:

"3.4. Model sensitivity as a function of the human acceleration of nutrient cycles
A detailed discussion of the model sensitivity for the year 2000 has been presented in Beusen et al. (2015). Here we focus on the impact of the acceleration of nutrient cycles during the

twentieth century on the model's sensitivity to changes in parameter values. Most parameters were varied within an interval of ± 10% around the default value. We consider parameters to have an important influence when they are significant for global delivery, retention or river export, and in addition, they exert a variation >4% of the default model (Tables SI4 for N and SI5 for P).

The influence of the natural ecosystem N budget on N delivery decreased from 1900 onwards (SRC decrease from 0.38 to 0.20) and was only important (0.21) for river N export in 1900. Likewise, allochthonous organic matter input was more important for N delivery in 1900 and 1950 than in 2000. It even exerts an important yet decreasing influence on river N export throughout the 20th century. P from allochthonous organic matter inputs was important for delivery and river export in 1900 (SRC = 0.23-0.24) while it was less important in 1950 and 2000. Weathering was important for P delivery in 1900 (SRC = 0.27) and 1950 (SRC = 0.23), and for river export in 1900 (0.21). P weathering is no longer important in the year 2000 due to the increasing delivery of P from fertilized fields and grazing land, and wastewater.

With a much smaller human population, less food and energy production in 1900 and 1950, the situation was different from the year 2000. Runoff had a smaller influence in the first half of the twentieth century than in 2000. Apparently, surface runoff was an important process for nutrient mobilization through leaching (N), surface runoff (N and P) and weathering (P) throughout the century. The influence of the agricultural N budget has been growing and became important in 2000, when its influence on N delivery to streams (SRC = 0.26) exceeded that of the N budget in natural ecosystems (SRC = 0.20). The influence of the P budget in agricultural fields has also been growing but remained an unimportant factor (SRC values < 0.20). The influence of the factors involved in the computation of P erosion (bulk density and P content of topsoil) was large in all years (SRC change from -0.54 to -0.63 for bulk density and 0.55 to 0.63 for P content between 1900 and 2000). This influence has been growing due to the increasing P inputs (fertilizer, manure) which partly determine P surface runoff, and due to the accumulation of P in agricultural soils in many world regions, particularly during the second half of the twentieth century.

The influence of N and P discharge from wastewater on the global scale was small (SRC >0.2 only for P discharge in 2000) compared to other anthropogenic sources such as agriculture. The data show that with smaller population densities in 1900 and 1950 as compared to 2000, wastewater also exerted a smaller influence on the delivery of both N and P in the first half of the twentieth century. Finally, temperature has a large influence on in-stream retention (SRC values of 0.34-0.41 for N and 0.21-0.27 for P) and river N export (SRC values of 0.30-0.36)."

10. Page 20136, Lines 7-9: Do you mean that desorption processes are not considered at all? not even as a percentage? Do the authors think that including desorption processes would change much the P export values to the sea?

Response: it is difficult to speculate about the impact of including this process in our model. Probably it would depend on the conditions such as the P saturation of the sediment, concentration in the water column, and the water flow. This is high on our list of processes to study. Nevertheless, to do so we need to build a more mechanistic model. We feel that this topic is highly speculative and have thus decided against discussing it in the manuscript.