Response to the comments about the submitted paper *"LPJmL-Med – Modelling the dynamics of the land-sea nutrient transfer over the Mediterranean region–version 1: Model description and evaluation"*

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We would like to thank Reviewer 1 for taking the time and effort necessary to review the manuscript. We sincerely appreciate all valuable comments and suggestions, which helped us to improve the quality of the manuscript (ms hereafter).

Please note that reviewers' comments are in blue while our answers are in black.

Reviewer R1

First of all I realize that the authors put a huge amount of work in the implementation and in writing this lengthy paper. This article tries to document the modeling concept,but this paper does not contain the quality needed for publication. There are a lot of reasons for this (see below). Beside that the structure of the paper could be improved. I had to go back and forth in this paper the whole time. It took me some time to make a review of this article. The formulas presented in this article are not new. The assumptions and the generation should be new. But on this part, I have a lot of questions or remarks. I stopped with remarks on typos and so on, because I believe this article needs a major revision before detailed feedback can be given.

We would like to thank the reviewer for highlighting the main points that should be considered under revision. The reviewer is right, this paper is the fruit of a considerable effort, starting from the implementation of the biogeochemical land-sea nutrient transfer processes within the LPJmL model, followed by the preparation of the inputs/boundary conditions of the model which required the manipulation of a large number of database, the calibration/evaluation of the model, and finally the writing of the paper.

We are aware that our approach still suffers from a lot of limitations and that the structure of the paper could be improved, but it intends to be a first proof of concept of the modelling of the transfer of nutrients from land to oceans. We hope that the corrections we brought based on the relevant advice from the reviewer help to clarify and improve the present paper.

Where is the NH4 in the rivers? Why leave this part of the dissolved N out of this paper? This is needed here as well.

As shown in the paper (section 2), NH4 is also implemented in LPJmL-Med represented through its dynamics in soil (equation 9) and in water (equation 13). Transformations between different forms of N in the soil are represented by remineralization, nitrification, and denitrification and are simulated in sequential order. We did not show the results of NH4, because in-situ data covering the whole Mediterranean basin are not available, which limits the evaluation of NH4 output, and on the other hand, the Mediterranean Sea is an exception with a strong P limitation (where N is generally considered to limit primary productivity in most of the world's oceans). For these reasons, and because of the greater availability of data on NO3 and PO4, we decided to focus hereafter on these two macro-nutrients.

However, as suggested by the reviewer, a figure of NH4 results has been added in the appendix of the paper (see the Fig. R1 above, see Figure A1 in the revised manuscript). In addition, a sentence has been added to the text to make this point perfectly clear in the revised manuscript (see sections 1 and 5.2.1).

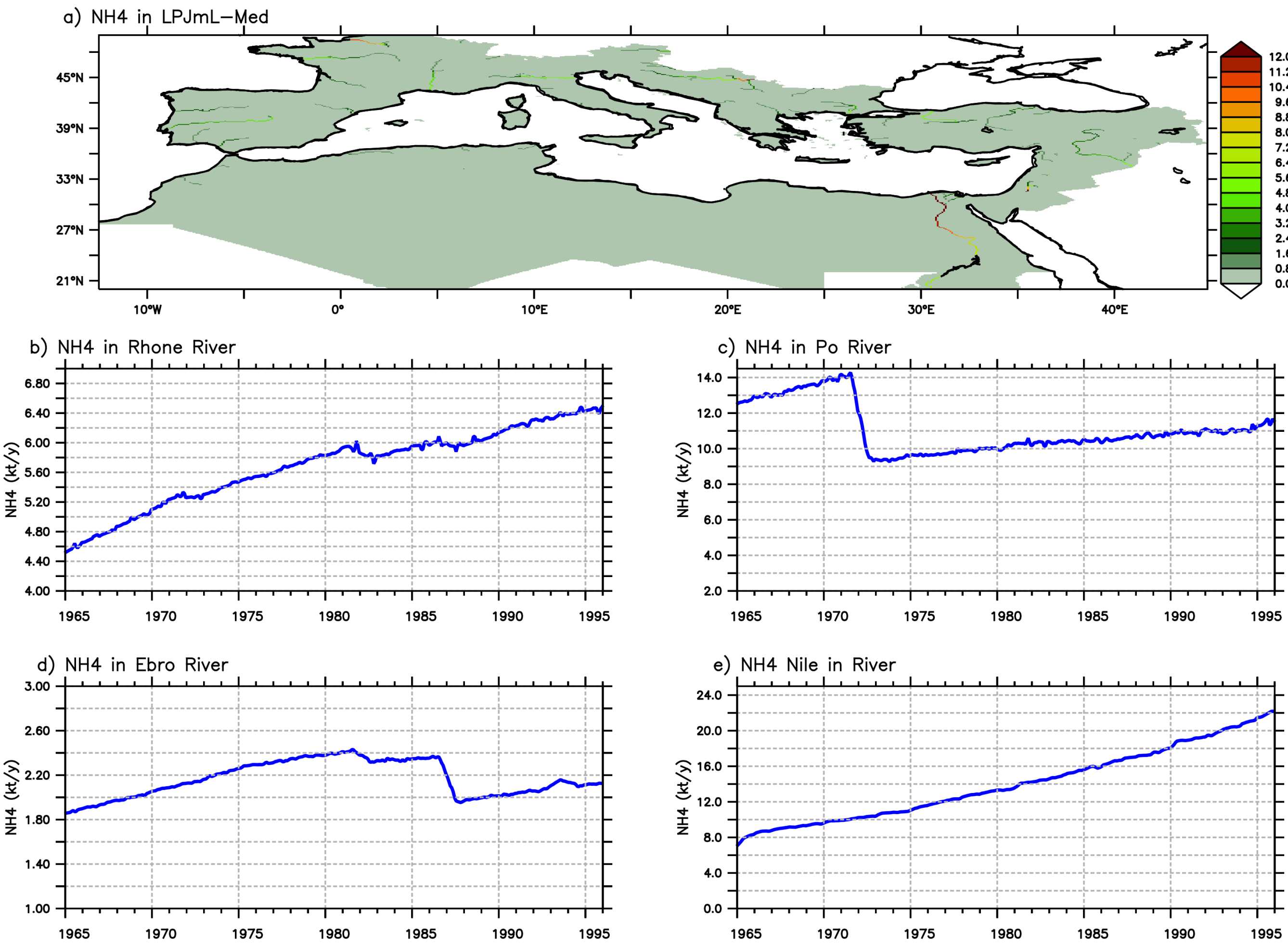

Figure R1: Annual NH_4 simulated by LPJm-Med in kt.y^{-1} flowing from the mains rivers into the Mediterranean Sea. **a**) horizontal map averaged between 1963 and 2005. (**b**, **c**, and **d** time series for the Rhone (station Beaucaire latitude: 43.92, longitude: 4.67), Po (station Pontelagoscuro latitude: 44.93, longitude: 11.52), Ebro (station Ortosa latitude: 40.82, longitude: 0.51), and Nile (average value of Nile mouth) rivers successively.

Soil and water temperature are needed in several equations. Where is their description?

Indeed, soil and/or water temperatures are needed in several equations. The description of the temperature parametrisation in the LPJmL model is fully detailed in Schaphoff et al.(2018, equations 19-25). However, the temperature-dependence is given in the present paper by equation

28 (cf. Line 340 in the submitted manuscript) describing a bell-shaped temperature dependence of each process considered (i.e. the difference between the optimal temperature and the environment temperature). We used monthly climate inputs data (i.e. precipitation and temperature data) from a simulation provided by the regional climate model CNRM-ALADIN (Herrmann et al., 2011; Farda et al., 2010; Déqué and Somot, 2008) and run in the framework of Med-CORDEX program (https://www.medcordex.eu).

Furthermore, a sentence has been added in the revised manuscript to improve the description of the temperature-dependence in the model (cf. below).

The bell shaped temperature-dependence of biological processes is driven by the difference between the optimal temperature and the environment temperature:

$$S(T_{opt}, \gamma) = exp\Big(- \frac{(T_{env} - T_{opt})^2}{\gamma^2}\Big) \tag{1}$$

where T_{opt} *is the optimal temperature for the process considered,* γ *is the sigmoid range of* T*, and* T_{env} *is the environment temperature (i.e. soil or water temperature depending on the process considered). The description of temperature parametrisation in the LPJmL model is fully detailed in [Schaphoff et al., 2018] (eq. 19-25).*

Where is the description of the organic part of P and N in the soils?

Only the detrital part of the organic N and P in the soils is explicitly represented in the model by the generic state variable $\mathrm{RES}_{X,l}$ where X either stands for N or P and l is the index of the soil layer. The equation describing the dynamics of these state variables is equation (2) of the submitted ms. The soil residue pool is fueled by the organic matter either coming from the dead biomass entering the litter pool or from the applied manure. The residue pool is updated for each soil layer, at different time steps depending on the source of the organic matter. As it is composed of organic matter, manure is added to the litter (see Fig. 1), and we distinguish between pastures and other crops (see equation 16). Residues added to the first soil layer (l = 0) correspond to the roots in that layer, as well as to dead organic material from the above-ground parts of the plants (see equations 26). The remineralization of N and P following the hydrolysis of organic N and P by soil decomposers is presented in equation 27. A sentence has been added to the text to make this perfectly clear in the revised manuscript (see description after equation 4 and Figure 1 representing the conceptual diagram of the model).

I miss an overview of the inputs, the delivery to the rivers, the in-stream removal and the export to the sea.

We are not sure to fully understand this question and assume that the referee is talking about an overview of **nutrients** inputs.

Inputs of nutrients:

The compilation of the data sets used in the LPJmL model for the inputs of fertilizer, manure and wastewater nutrients content [1961-2005] are listed in section 4.1. In the beginning of section

4.1, we explain that this section will describe all model inputs, including the nutrients load from agriculture and wastewater release, especially quantified for the present model development. Moreover, Table A2 in the submitted ms presents an overview of all the inputs used in the simulation.

Transport of nutrients:

The transport of nutrients along water flow is performed through river-routing as shown in section 3.2. Water content in soil layers is altered by infiltrating rainfall, gravity (percolation), and by plant uptake. The infiltration rate of rain and irrigation water into the soil depends on the current soil water content of the first layer. The water surplus which does not infiltrate is assumed to generate surface run-off.

The lateral exchange of water discharge between grid cells through the river is computed via the river-routing module implemented in LPJmL-Med model. The transport of water in the river channel is approximated by a cascade of linear reservoirs [Schaphoff et al., 2018, Rost et al., 2008]. Cells receive water from the reservoirs when the following conditions are met: (i) cells have a lower altitude than the cell containing the reservoir, and (ii) they are located along the main river downstream or at maximum five cells upstream [Schaphoff et al., 2018]. Hence, a cell can receive water from multiple reservoirs. Both surface and subsurface run-off are simulated to accumulate to river discharge (for more detail see [Schaphoff et al., 2018]).

Removal and export: All processes leading to a loss of nutrients from soils and rivers (i.e. sedimentation, adsorption and denitrification) are presented in section 2.4. Part of the inorganic nutrients get adsorbed on particles and are lost through sedimentation, both in soils and rivers (e.g. [Thieu et al., 2009]). Adsorption only concerns PO_4 due to its chemical properties, it has an important role in determining the relative proportion of nutrients in both terrestrial and aquatic systems. The daily amounts of PO_4 adsorbed on soil particles ($ADS_{PO4,l}$), is computed from equation 34, cf. section 2.4.3 in the paper.

The description of inputs and the delivery to the rivers has been improved in the revised ms see section 4.1.1.

Validation of discharge, NO3 and PO4 should be done on the same time scale. It is very confusing. Why only to the year 2000? That is still 20 years back from now!

In this study, we focus on the historical period of the simulation, namely the 1963-2005 period. From 2006, the climatic forcing (provided by a simulation run with the CNRM-ALADIN model in the frame of the Cordex program), and therefore the simulations presented in the present work, switch to a RCP scenario. To clarify this point, a new figure R5 presenting the different simulations run in this study has been added in the revised ms (see the new Fig. 2b in the revised ms). Moreover, the time scale of Figures 9 and 10 are now the same and encompass all the historical period (i.e. from 1963 to 2005), as suggested by the referee (cf. Fig. R2).

Questions

Line 123-124: State variables: N and P contents has a fixed C:N and C:P ratios? Very strange. Why should this be?

Homeostatic regulation of microbial biomass composition constitutes the basis for the consumer-driven nutrient recycling theory [Sterner and Elser, 2002, Elser and Urabe, 1999, Sterner and Elser, 2002], according to which the elemental ratios of consumers and their resources determine the ratio of

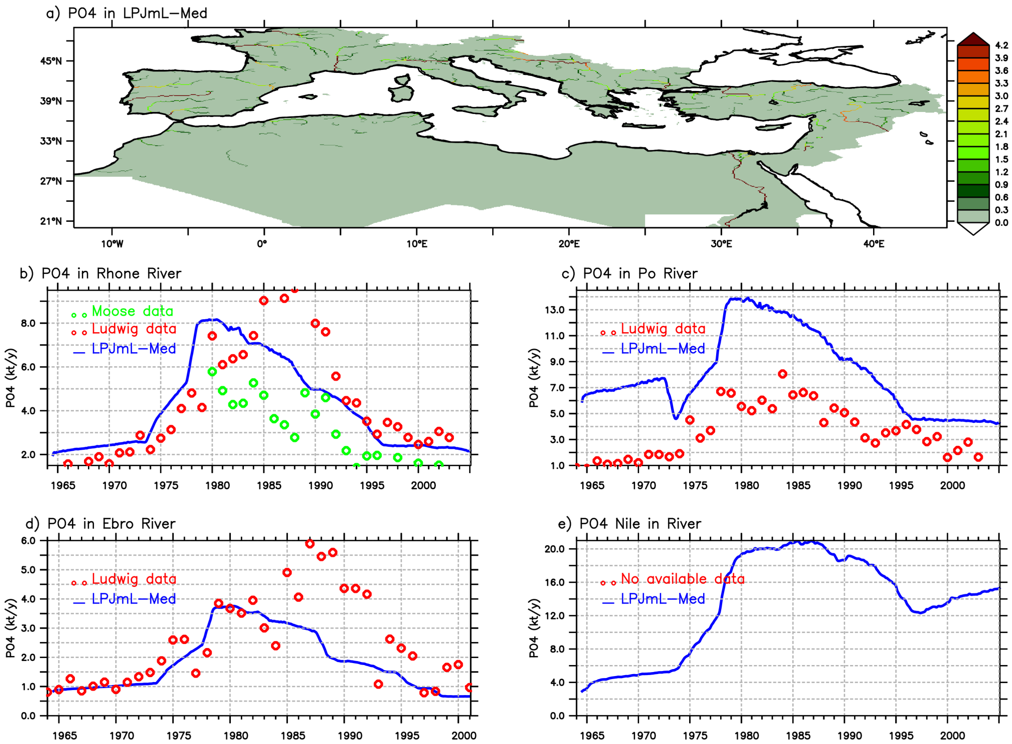

Figure R2: Annual PO_4 simulated by LPJm-Med in kt.y^{-1} flowing from the mains rivers into the Mediterranean Sea. **a**) horizontal map averaged between 1963 and 2000. (**b**, **c**, **d**, and **e** time series for the Rhone (station Beaucaire latitude: 43.92, longitude: 4.67), Po (station Pontelagoscuro latitude: 44.93, longitude: 11.52), Ebro (station Ortosa latitude: 40.82, longitude: 0.51), and Nile rivers (average value of Nile mouth) successively. Model output in blue, in-situ data (from Ludwig2009 Ludwig2009) in red, and in green from MOOSE program from [Raimbault and Lagadec, 2012]

C:nutrient released through differential recycling of C and nutrients (N or P).

However, these processes are still insufficiently understood and documented so far, which led us to consider in the model that the N and P content of decomposers in the soil layer are inferred from the carbon content of decomposers using fixed C:N and C:P ratios. The latter ratios have been proposed by [Mooshammer et al., 2014] in which they synthesize the current state-of-the-art of microbial adaptations to resource stoichiometry, and they estimate the C:N and C:P ratios of a selection of plants. Thus, a deeper understanding of the interactions between heterotrophic microbial communities and their chemical environment remain a key questions to better understand the functioning of the terrestrial ecosystems and this is indeed a direction for future improvements. A sentence has been added to the text to make this perfectly clear in the revised manuscript :

"The latter ratios have been taken from the review proposed by [Mooshammer et al., 2014] in which they synthesize the current state-of-the-art of microbial adaptations to resource stoichiometry, and they estimate the the C:N and C:P ratios of a selection of plant (i.e. roots, sapwood, leaves, storage)". See section 2.2 in the revised ms.

Equation 2. Here MAN_X is used, but the description says it is organic matter. So I miss this components in the description of N and P. The organic P storage in the soil behaves totally different than the dissolved P storage. Then equation 4 shows a constant ratio C:P which I really doubt. The storage of P in the soils works different than the storage of C.

MAN_X is the content in element X (X= N, P) of the daily applied manure (section 2.3.2). As it is composed of organic matter, manure is added to the litter (see Fig. 1). To estimate the amount of N manure introduced in each stand (MAN_N), on days when this occurs, we distinguish between pastures and other crops (cf equation 16). As data for P manure application are not available from the FAOSTAT database, we estimate it using a constant country-specific (P_{MAN} : N_{MAN}) ratio provided by [Potter et al., 2011] (cf. equation 17). N:P ratios in manure derived from [Potter et al., 2011] are used to estimate P manure applied to cultivated soils or left on pasture.

Unlike fertilizers providing mineral N and P nutrients, manure provides organic nutrients that are not immediately available for the plants, therefore manure is applied earlier in the year than the synthetic fertilizers. The equations used for calculating the annual nitrogen and phosphorus content of manure are described in Section 2.3.2. All the processes leading either to a change in the form of nutrients (i.e. primary production, residue decomposition and nitrification) are presented in section 2.4.

Equation 10: Which part of P fertilizer is available for uptake by plants? Why is the REMIN-PO4 multiplied by PO4. Should that not be organic-P?

We apologize for this oversight on equation 10, this was a typo. We have now corrected this equation in the revised ms as follows:

$$\frac{\partial NH4_l}{\partial t} = FERT_{NH4,l} + REMIN_{NH4,l} \cdot RES_{N,l} - UPT_{NH4,l} - NITR_{NH4,l} + LEACH_{NH4,l} \quad (2)$$

$$\frac{\partial PO4_l}{\partial t} = FERT_{PO4,l} + REMIN_{PO4,l} \cdot RES_{P,l} - UPT_{PO4,l} - ADS_{PO4,l} + LEACH_{PO4,l} \quad (3)$$

where $\text{RES}_{X,l}$ is the residue content in the soil layer, and $REMIN_{X,l} \cdot RES_{X,l}$ represents the source of nutrients from the remineralization following the decomposition of organic P (from manure and litter residues, sections 2.3.2 and 2.4.1).

Fertilizer only on crop fields. In Europe fertilizer is also used on grassland.

Yes, fertilizers are very intensively used in some European grasslands, especially in northwestern Europe, but not much in Mediterranean Europe. [Porqueddu et al., 2016] stated that "Due to physical and climatic constraints, Mediterranean grassland systems are usually extensive, with low usage of pesticides, fertilizers, concentrates and irrigation". We acknowledge that the model is also run for areas with a lot of grasslands under oceanic climate that are fertilized (like e.g. western France), but apart a few exceptions (upper Rhone River catchment areas) these areas do not belong to the Mediterranean catchment area, therefore they do not impact the results discussed in this paper. Generally, Mediterranean grasslands receive much more manure than synthetic fertilizers (see e.g. [Bouwman et al., 2002]), so we decided to consider only manure fertilization for grasslands in this study.

Equation 11: Why is growth of phytoplankton not dependent on NO3 and/or PO4? It turns out that in equation 30 this is not true. Why not moving equation 11 to 2.4.2? I need a lot of searching to find the explanation for some of the processes. Is there another structure of the description possible?

We have chosen this organisation of the paper so as the paper can be read at two levels. The first level addresses readers that are only interested in knowing the nature of the processes that are included in the model and the corresponding mass fluxes and they can just focus on the general conservation equations given for each state variable at the beginning of the paper without being caught up in the details. The second level consists of all the details that are given process by process in dedicated sections. We believe that this organisation of the paper is the least worst solution.

Concerning the question about phytoplankton growth, the latter indeed depends on NO3 + NH4 and/or PO4 (as shown in equation 30 of the submitted ms), and on temperature (using bell shaped temperature-dependence between 14 and 24 °C). Phytoplankton can become limited by the availability of nutrients when light and temperature are adequate and loss rates are not excessive.

Line 207-211: Nice to include this remark, but assumption of N-fixers is not correct, and excluding atmospheric deposition (land and water) is a pitty. So both should be included!

We strongly agree with the reviewer that atmospheric deposition (land and water) is potentially an important source of nitrogen and phosphorus. However, the existing time series atmospheric deposition data cover only very limited areas of the basin and short time periods. Moreover, there is, to our knowledge, very few experimental study addressing the source apportionment of phosphate deposition [Longo et al., 2014, Desboeufs et al., 2018], and the bioavailability of these atmospheric inputs is not fully understood.

We underline here the need for more deposition measurements in order to better constrain the modeling of such important nutrient sources for the Mediterranean. Further development of atmospheric and oceanic models should be undertaken in order to account for the mixing and chemical processing of the different aerosol sources in the atmosphere and their effect on nutrients solubility in water. A sentence has been added to the text to make this perfectly clear in the revised manuscript (see section 2.2).

Biological nitrogen fixation is an important natural process in which N2-fixing bacteria (diazotrophs) convert atmospheric nitrogen into plant-usable forms such as ammonium and nitrate. Currently, there is very limited information and data on the importance of this process in the Mediterranean region.

Why is 50% NO3 and 50% NH4 of fertilizer? This is dependent of the type of fertilizer. It is a pitty when you consider NH4 and NO3 and just divide N into two.... Some type of fertilizer contains a lot of NH4, some don't. Please improve.

The IFADATA database [IFA, 2016] provides N and P fertilizer data without giving the details of the allocation of N into NO3 and NH4 pools. According to the literature, ammonium nitrate is the most used N fertilizer in Mediterranean countries (see e.g. [Isherwood, 2010, Ryan et al., 2009, Gezerman, 2020]). Ammonium nitrate is made of ammonium and nitrate in equal amount, therefore we assume that half of the inorganic N inputs from fertilizers enters the nitrate pool in the first soil layer and the remaining half feeds the ammonium pool also in the first soil layer. A sentence has been added to the text to make this perfectly clear in the revised manuscript. See section 4.1.1 in the revised ms.

Line 253: What is cultivated land? Cropland and grassland?

We agree with the reviewer that the word of "cultivated land" was not very clear, and we have changed by agricultural land in the revised ms that encompasses cropland (annual and perennial)and managed grassland (mown or pastured).

Equation 15: Why area weighed distribution and not crop weighed. With LPJmL you have different crop types, so make better use of this information!

We agree with the reviewer that it would be better to consider crop-specific fertilizer distribution, however such data are not available under the form of time series for all the countries as this is needed by the model. The yearly N and P fertilizer consumption data provided by the international fertilizer association [IFA, 2016] are country-specific but not crop-specific, therefore the fertilizers area-weighted distributed, i.e. considering the cropland area (pulses excluded) of each cell.

Equation 17: Values of Pman and Nman are not given. Why take a constant ratio? It is dependent on the type of animals that produce the manure. So it is country specific and time dependent.

P_{manure} application data are not available from the FAOSTAT database and we estimate it using a constant country-specific ratio provided by [Potter et al., 2011]. Potter et al. calculated spatially explicit fertilizer inputs of nitrogen (N) and phosphorus (P) by merging national-level statistics on fertilizer use with global maps of harvested area for 175 crops. They also calculated spatially explicit manure inputs of N and P by merging global maps of animal density and international data on manure production and nutrient content. Finally, manure data are indeed country specific and time dependent as shown in figure 4.

Line 251: Again the authors assume that 50% of sewage effluent is nitrate and 50% is NH4. Why? This is not a reasonable assumption!

We thank the referee for this very relevant comment. We now assume in the revised version that 99% of sewage effluent is NH4, as reported in many sources. We have re-run all the simulations using this more realistic value of the ratio of NH4 in sewage effluent. Figure R3 shows the new results of the model (LPJmL revised version). This new version of the model improves the simulation outputs of NO_3 flowing from the mains rivers into the Mediterranean Sea as compared to available in-situ data and to the previous version of LPJmL-med model (cf. Figure R3). Corresponding changes in the revised ms are given in section 2.3.3.

Line 255. Where is the input from industry?

We used the formulation of [Van Drecht et al., 2009] to calculate N content in waste water. Despite writing in their article that human N emission is the N emitted in wastewater by households and industries (connected to the same sewerage system), the authors actually only consider the "households" human emissions of the population connected to public sewerage systems (see equa-

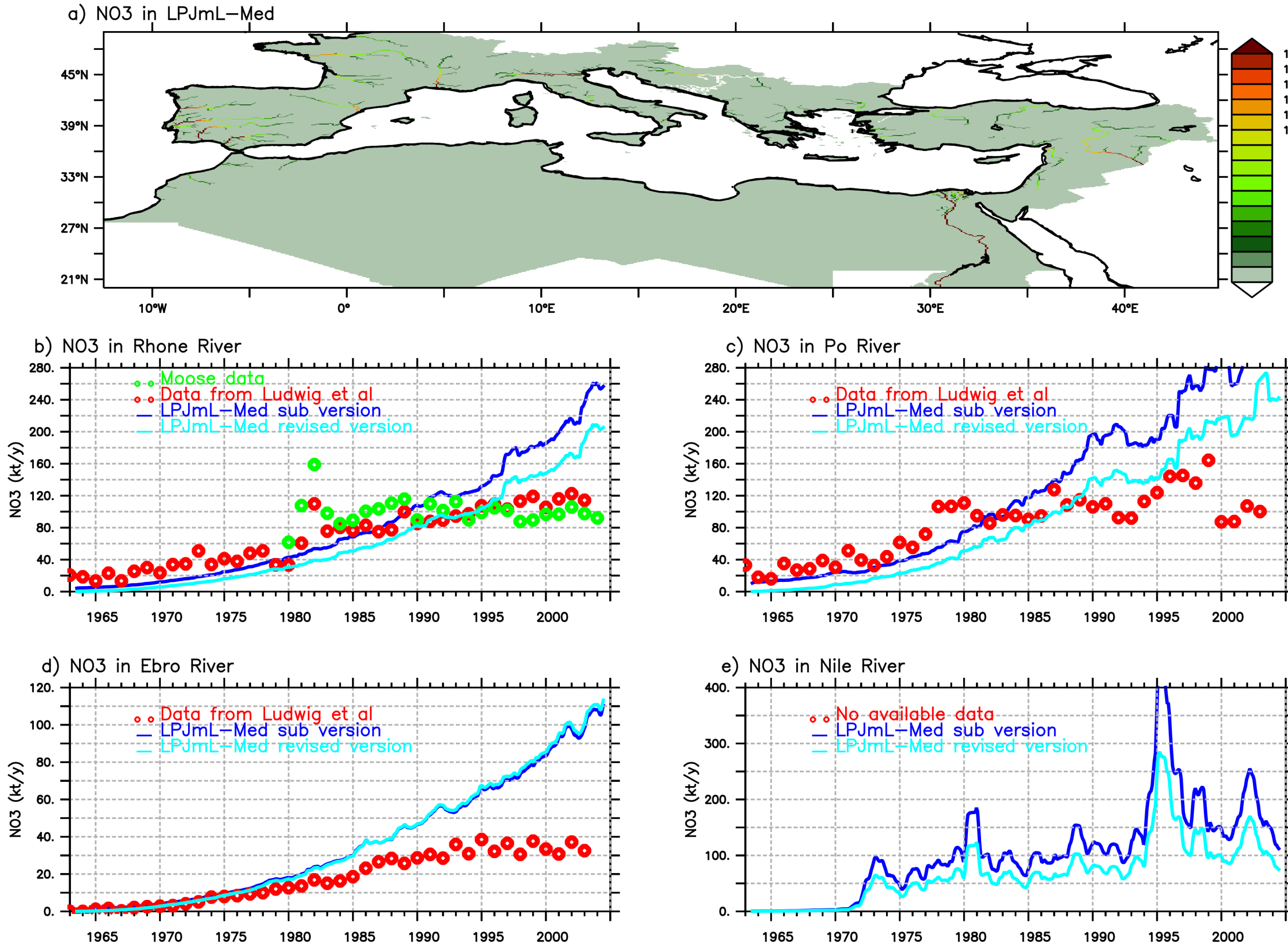

Figure R3: Annual NO_3 simulated by LPJm-Med in kt.y^{-1} flowing from the mains rivers into the Mediterranean Sea. **a**) horizontal map averaged between 1963 and 2000.**b**, **c**, **d**, and **e** time series for the Rhone (station Beaucaire latitude: 43.92, longitude: 4.67), Po (station Pontelagoscuro latitude: 44.93, longitude: 11.52), Ebro (station Ortosa latitude: 40.82, longitude: 0.51), and Nile rivers (average value of Nile mouth). Model output in blue, in-situ data (from [Ludwig et al., 2009]) in red, and in green from MOOSE program from [Raimbault and Lagadec, 2012]

tion 1 in [Van Drecht et al., 2009]. In the conclusion, they acknowledge that main uncertainties concern industrial emissions, which are not explicitly included in their approach.

I am surprised by taking the waste water model of [Van Drecht et al., 2009]. The problem is the assumptions of the GDPppp and GDPmer which is from US dollars of 1995. There exist almost no model which uses this specific GDPppp and GDPmer. So the authors will run into problems when applying this model. Besides there are some updates of this model [Morée et al., 2013, van Puijenbroek et al., 2018]. Equations 21-24 are dependent on this GDP.
We do not know all waste water models, but we found quite a lot of studies that have applied the model of [Van Drecht et al., 2009] for computing point sources nutrient inputs in different nutrient export models (NEW, NEW 2, MARINA, etc). We can cite [Mayorga et al., 2010,

Sattar et al., 2014, Strokal, 2013, Pedde et al., 2017, Wang et al., 2019], which all rely on GDPppp and GDPmer following [Van Drecht et al., 2009]. The World Development Indicators database provides data about the interannual variations of the US dollars consumer price index, allowing to compute any GDPppp expressed in US dollar for any year in the ppp-based GDP in 1995 US dollars as used in [Van Drecht et al., 2009]. For the present paper, we therefore consider this model as an usual and accepted methodology. Indeed this model has been updated, and we plan to use the formulations of [van Puijenbroek et al., 2018] for future work when applying our model under climate and socio-economic change scenarios.

Line 272: I don't think this ratio is the N:P ratio of municipal wastewater. This is the human intake. I miss for a lot of equations the units (for example equations 21-24).

The referee is right, this is the human intake, representing the ratio between human P and human N emission (no dimension). We use the standard value of 6 derived from measurements of 27 wastewater treatment plants in Austria [Van Drecht et al., 2009, Zessner et al., 2005]. This has been corrected in the revised ms, see description after equation 21.

In this paper, we have more than 45 equations and many variables, and as much units. Hence we have putted the units/value of all parameters used in the model in Tables A1, A2, and A3. All state variables and their units are presented at the beginning of section 2.2.

The C-susp-water is calculated (equation 36). But this is only dependent on land-use. So the amount of suspend matter in the river is independent on the surface runoff or land erosion. Is this assumption valid?

We acknowledge that we should have better explained this equation. Equation 36 gives the concentration of suspended particles in water. The values used in equation 36 (given in Table A1) are the concentrations of suspended matter at the surface runoff for the different land-use classes as provided by [Billen and Garnier, 2007] in their Table 2a. The concentration of suspended matter in the surface runoff directly depends on the land-use. Erosion processes produce the particles of solid matter, that will be transported by the runoff flow. Therefore, the coefficient used in equation 36, that was empirically obtained, implicitly accounts for erosion. It would be better to realistically represent erosion processes in the LPJmL model to account for the role of soil type, slopes, climate, vegetation of the riparian zone, in addition to land use for estimating the amount of rill erosion that will be responsible for the particulate matters. But LPJmL does not simulate erosion. Due to the low vegetation cover and the importance of sloppy areas, many Mediterranean areas are indeed vulnerable to erosion. However, [Cerdan et al., 2010] showed that erosion rates are much lower in the Mediterranean than in the rest of Europe due to higher presence of rock fragments and quasi-absence of highly erodible loess soils. Following and without a proper erosion model, we work with the hypothesis that we can use the land use specific concentrations of suspended particles in water as provided by [Billen and Garnier, 2007].

Line 442: So maximal depth of soil is 3 metres. So there is no groundwater modelled here? Also in equations 45-49, I don't see any delay in nitrate going through the soil layers. Can you elaborate on this?

Yes, in the current version of LPJmL-Med the ground water is no simulated and we are aware that this is a strong limitation of the model and a potential direction for future work. Concerning the delay in nitrate going through the soil layers, it is implicitly considered through the amount of water percolating to the underlying soil layer per day since the nitrate flux is proportional to this water percolating flux. For this, we have used the existing parameterisation of LPJmL5.0 version published by [Von Bloh et al., 2018] and based on the method used in SWAT model [Neitsch et al., 2005, Neitsch et al., 2002].

Line 512: missing ares?

Sorry, this was a typo. We meant "the missing are**a**".

Line 535: I read that legumes don't get any N and P fertilizer?? Strange.

Due to their ability to fix atmospheric N, legumes require almost no N fertilizer [Stagnari et al., 2017]. On the other hand, they benefit from P fertilizers, but observations generally report less P fertilizers use for legumes than for cereals (see e.g. [Reckling et al., 2014]). Unfortunately, the database on Crop Specific Fertilizer Rates [Müller et al., 2012] does not consider legumes, and the reports from the International Fertilizer Association mix legumes with other crops (e.g. [Heffer, 2013]). Considering the fact that, in southern Europe, the organic fraction of legumes areas (i.e. without any synthetic fertilizer) is higher than that of any other crops (AgenceBio 2019), and that legumes area remain low, we decided to not distribute fertilizers on legumes. A sentence has been added to the text to make this perfectly clear in the revised manuscript (see section 4.1.1).

Line 537: Is there a fertilizer application of grass or not?

No synthetic fertilizers are not applied on grassland, only manure (i.e. organic fertilization) is applied on grassland. This is now clearly stated in the revised ms, see section 4.1.1.

Figure 3: typos in headers, What does c and d contain? Load in the main stream of the rivers? Where? At the mouth of the river? What is the unit? In the text it is always PO4-P and NO3-N. But here in the figures? Make clear. How is it possible that PO4 is twice as high as NO3 (c and d)? Why is y-axes of f three times lower than d? The sum of three rivers (c) is higher than sum of two regions. Why? Is a and b fertilizer on the soil? Is NO3 fertilizer in a half of total fertilizer?

We apologize for this oversight on Figure 3, the caption was uncomplete. The figure will be re-plotted following the advice of the reviewer (see the new figure R4 below).

What does c and d contain?:

They contain the time series of yearly averaged values of N-NO3 and P-PO4 (in kilotons) of fertilizer at the same locations as river gauging stations:

Rhone: Station Beaucaire (France, latitude: 43.92, longitude: 4.67).

Po: Station Pontelagoscuro (Italy, latitude: 44.93, longitude: 11.52).

Ebro: Station Ortosa (Spain, latitude: 40.82, longitude: 0.51).

In the text it is always PO4$-P$ and NO3$-NP$ But here in the figures?:

As noted in the text (see line 607), NO_3 refers to N-NO_3 and PO_4 to P-PO_4 (line 631). Annotations will be homogenized in the revised manuscript.

How is it possible that PO4 is twice as high as NO3 (c and d)

The use of chemical phosphorus (P) fertilizer was very important in Europe between 1970 and 1990 as shown in Oscar et al. (2015). All the data plotted in Figure 3 are downloaded from IFADATA dataset (IFA, 2016).

Why is y-axes of f three times lower than d?

Because we plotted the mean values in the eastern and western basins of the Mediterranean Sea in Fig. 3f. This has been changed in the revised ms where we plotted instead the sum of Eastern and Western basins (cf. new Figure 3 in the revised ms, Fig.R4).

The sum of three rivers (c) is higher than sum of two regions. Why?

You mean by sum of two regions: eastern and western basin ?, in Fig. 3 we plotted the average value to compare the global trend in the two basins. It has been changed in the revised ms where we plotted instead the sum of the Eastern and Western basins (cf. new Figure 3 in the revised ms (Fig.R4).

Is a and b fertilizer on the soil

N and P fertilizers are applied to the first soil layer as explained in section 2.3.1 entitled *Fertilizers application*.

Is NO3 fertilizer in a half of total fertilizer? We used N and P fertilizer data provided by IFADATA dataset [IFA, 2016], we consider that half of the inorganic N inputs from fertilizers enters the nitrate pool in the first soil layer, and the remaining half feeds the ammonium pool (cf. section 2.3.1).

Figure 4: I don't understand this figure. Manure is fully organic matter. So NO3 and PO4 are zero. I don't know what is presented here.

The referee is right, the use of NO3 and PO4 is very confusing for the reader. In fact, we presented in Fig.4 the MAN_N and MAN_P as calculated in section 2.3.2. As it is composed of organic matter, manure is added to the litter (see Fig. 1). Figure 4 and the associated text were corrected in the revised manuscript.

Figure 5: I am completely lost. The unit here is in tonnes. Which means that WWT is very very very small compared to fertilizer or manure. Something is wrong.

The wastewater inputs are in tons, because wastewater is released directly into rivers and reservoirs as shown in Fig. 1 showing the N, P contents from sewer systems (wastewater release). It is indeed very small compared to fertilizer or manure, because the latter are added to soil and associated with agricultural practices (i.e., application of manure and inorganic fertilizers to soils). A sentence has been added to the text to make this perfectly clear in the revised manuscript, see the description of wastewater inputs in section 4.1.1.

Line 550 – 556: GDP is not used.

We are not sure to fully understand the question. Lines 550-556 explain the wastewater dependency on GDP, reporting to the Section 2.3.3 for the equations (21-24). These equations show that the per capita nutrient releases in waste wate (human emission, laundry, dishwasher) depend on GDP.

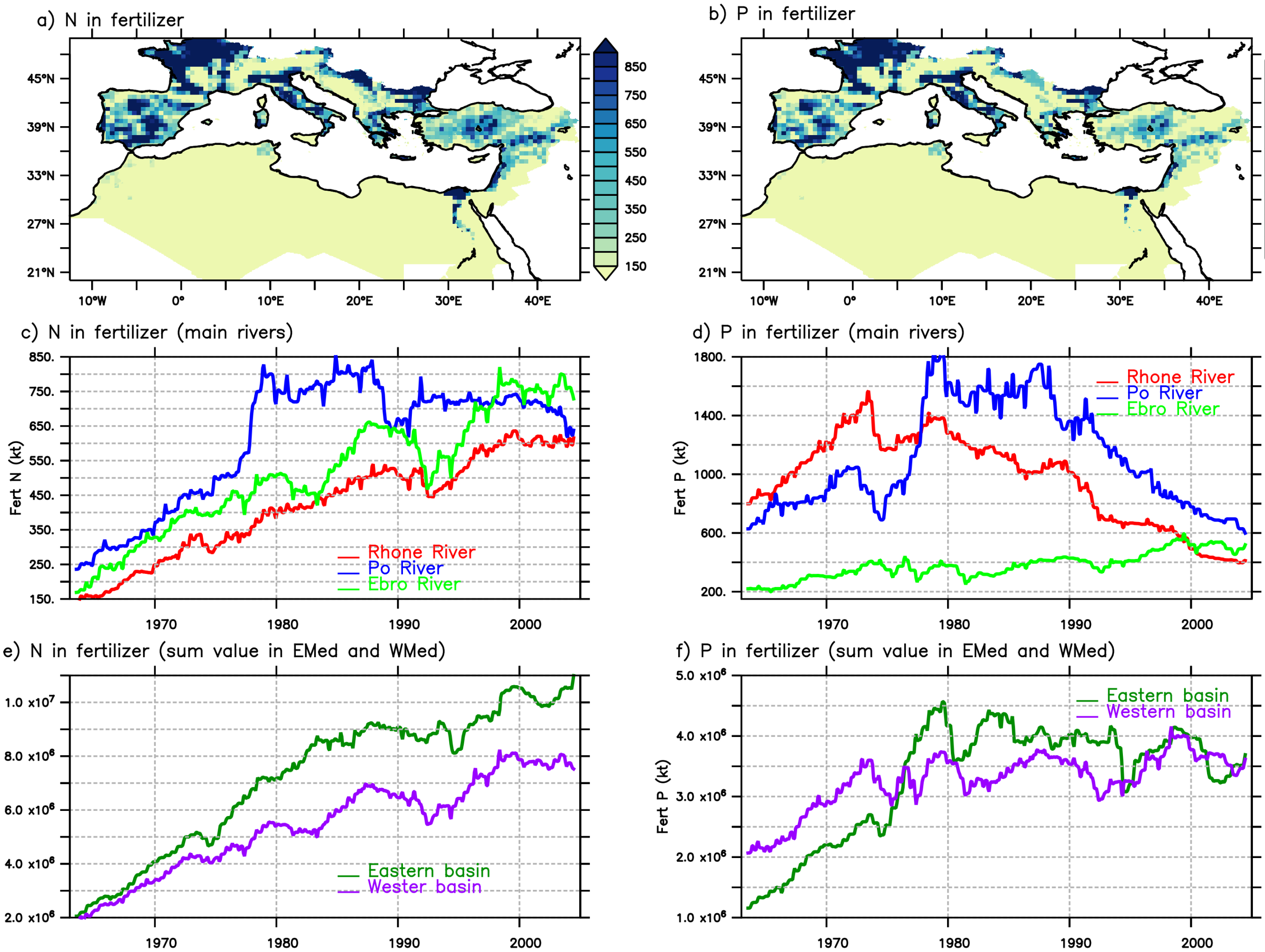

Figure R4: A compilation of nutrient inputs data set from fertilizer data (in kilotons/pixel of N and P), (**a** and **b**) horizontal maps represent LPJmL-Med annual input of fertilizers averaged between 1960 and 2005. (**c** and **d**) time series near the Rhone gauging station in red (station Beaucaire latitude: 43.92, longitude: 4.67), Po gauging station in blue (station Pontelagoscuro latitude: 44.93, longitude: 11.52) and Ebro gauging station in green (station Ortosa latitude: 40.82, longitude: 0.51). (**e** and **f**) times series for eastern and western basins.

Line 566 – 567: "In general . . .. is poorer" Why is this statement here?

Text changed to: "In general, the southern Mediterranean rivers are much less well documented compared to northern-European rivers. Particularly, long time series of data are missing for southern Mediterranean rivers."

Figure 6: Why is the Nile here? There is no comparison with observations. Why upper figure average of 1963–2000? Figure caption claims 1920 – 1985, that is only for Ebro.

We have added the Nile to all the all the figures of results because the Nile is very important river in the mediterranean region (i.e. among the longest river in the world). It is very difficult

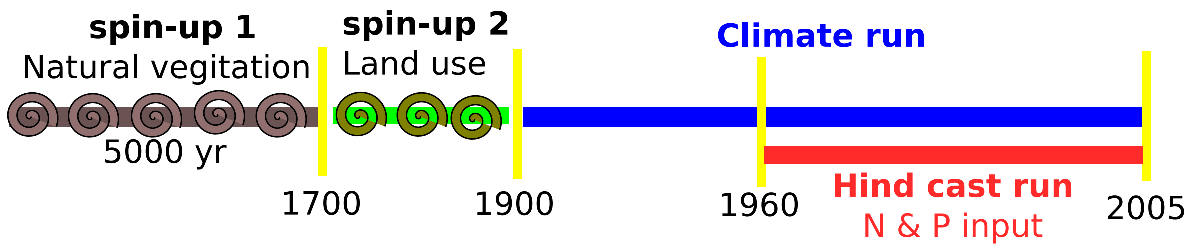

Figure R5: Presentation of the different simulations run in this study. We started by a 5000-year spin-up simulation with water and carbon pools set to zero at the beginning of the simulation to bring natural vegetation patterns and carbon stocks into dynamical balance. During this spin-up, 30 years of climate forcings with constant concentrations of atmospheric carbon dioxide (at 278 ppm) are periodically repeated. Then a second spin-up was run (during 500 years) during which land use is introduced in the year 1700 from which it is updated annually according to the historic land use data-set (see Fader2015, Fader2010). Finally we run a hind cast simulation [1960-2000] which includes the new feature developed in the present work, namely N and P cycles.

to evaluate and simulate the discharge from the Nile, because very few data are available, and the construction of the Aswan High Dam has had a major impact on the water discharge. Furthermore, we have evaluted the witer discharge of Nile river simulated by LPJmL-Med at the end of section 5.1.

LPJmL-Med simulates relatively well the water discharge of Nile with an average value of 17.88 $km^3.yr.^{-1}$ *(average value between 1965 and 1985), despite being relatively higher than the previous estimation from literature [ElElla, 1993, Nixon, 2003], but still far lower than the estimation of [Skliris et al., 2007] of about 83* $km^3.yr^{-1}$ *at Aswan.*

We agree with this comment on fig. 6. The different time periods could be very confusing for the reader. To clarify this point in the revised manuscript, we will add a new figure (see figure R5).

Why upper figure average of 1963–2000

To show an average state of historical water discharge in the Mediterranean Sea. A sentence has been added to the text to clarify this point in the revised manuscript.

Figure caption claims 1920 – 1985, that is only for Ebro

We only plotted the periods corresponding to available in-situ data; i.e. Rhone (1920 -1985), Po (1918-1989), Ebro (1910-195). However, for the sake of clarity, we will re-plot Figure 6 in the revised ms using the same period of time (i.e. 1920-2005) for all the rivers (cf. Figure R6).

Line 576: Why limit the impact of damming and anthropogenic water use? This is one of the things your model can do, so I don't understand this. Why mentioning "but not Nile"?

"limit the impact of damming and anthropogenic water use": We agree with the reviewer that this sentence was not very clear and could lead to a potential confusion. We therefore chose to remove this sentence from the text.

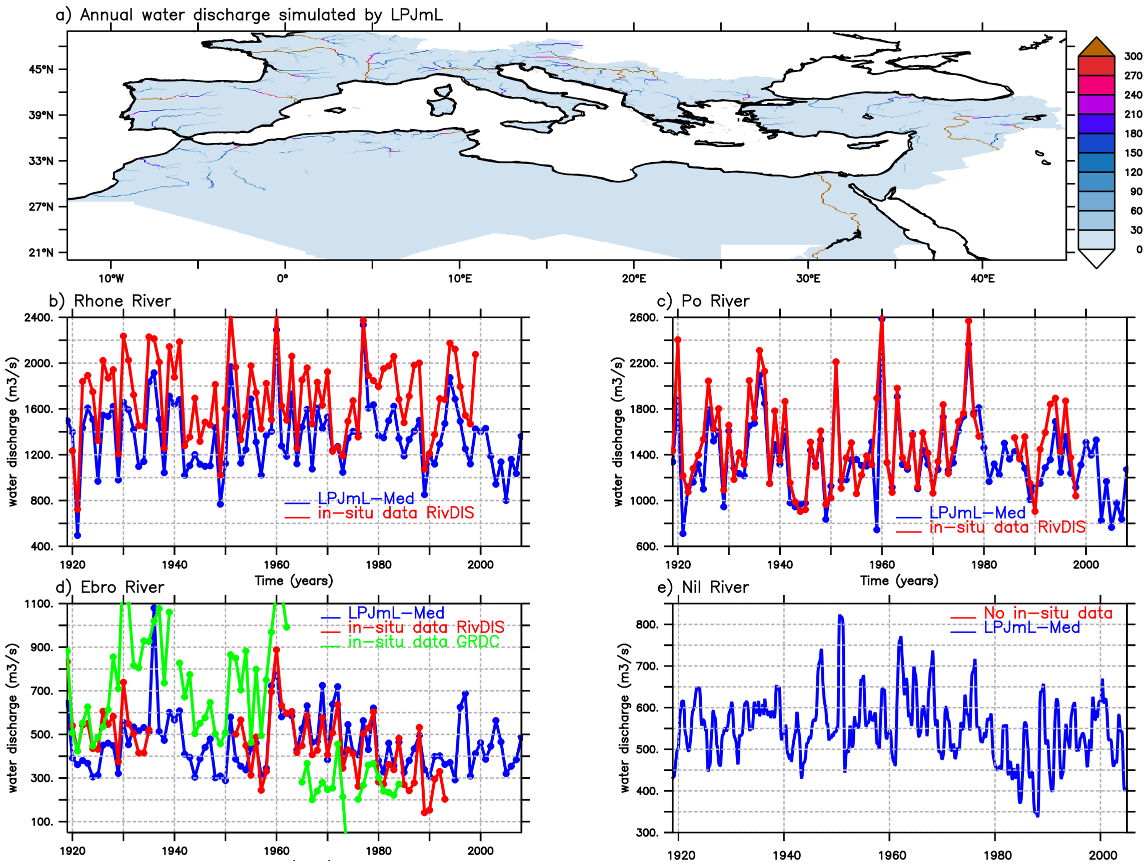

Figure R6: Yearly averaged water discharge in m^3/s for the main rivers into the Mediterranean Sea. **a**) horizontal map on implemented rivers in the LPJmL-Med grid (Averaged between 1963 and 2000). **b**, **c**, **d**, and **e** time series of water discharge at the mouths of Rhone (station Beaucaire latitude: 43.92, longitude: 4.67), Po station Pontelagoscuro latitude: 44.93, longitude: 11.52), Ebro station Ortosa latitude: 40.82, longitude: 0.51) and Nile (average value of Nile mouth), successively. Model output in blue, in-situ data (from the Global River Discharge database RivDIS [Vörösmarty et al., 1996]) in red, and from CFDC database in green.

" but not Nile" : We meant by this sentence that in-situ data are available for the main rivers (i.e. Rhone, Po, Ebro) except the Nile river where in-situ data are not available for a long period, and the construction of the Aswan High Dam has had a major impact on the water discharge of this river.

Yes, the LPJmL-Med model is able to simulate the impact of damming on rivers. However, this water regulation by dams is difficult to represent explicitly because very few data are available and those regulations are sporadic and hard to characterize, particularly for long periods. Hence, in this study we used a constant ratio to represent the impact of damming on the water use.

I think there is a misunderstanding in Figure 7. Here there is a comparison between modeled data and observed data. I miss the 1:1 line. What calibration data is used to calibrate the discharge of this model? At which location is this comparison? Why here 1920–1980 and not 1985?

We apologize for this missing, the caption was incomplete. This is not a calibration of the water discharge provided by the LPJmL-Med model but rather a comparison between measured and modelled discharges. Figure 7 indeed shows a scatter plot of the yearly water discharge from in-situ data for the Rhone (1920 -1985), Po (1918-1989), and Ebro (1910-1995) Rivers, and the water discharge calculated by the model for the same period are superimposed. We have improved the explanation in the revised version.

At which location is this comparison?

Again, we took the same locations as the ones of river gauging stations (the locations was added to all figure legends in the revised ms):

Rhone: station Beaucaire (France, latitude: 43.92, longitude: 4.67).

Po: station Pontelagoscuro (Italy, latitude: 44.93, longitude: 11.52).

Ebro: station Tortosa (Spain, latitude: 40.82, longitude: 0.51).

Line 586: Why the word “may”? Do you mean that Lutz and/or Ludwig is claiming this or are you seeing this in the model data?

Yes, it has been demonstrated by Ludwing et al., 2009, cf. section 3.2. in Ludwig et al., (2009), and Lutz et al., (2016) have confirmed the severe decline in streamflow for the Ebro.

As shown in figure 6 (from in-situ data and model results), the Ebro River shows a net decrease in water discharge between 1960 and 1990 (from 28 to 9 $km^3.y^{-1}$), due to the higher frequency of dry periods observed in the Ebro River basin during the same period.

However, the Po and the Rhone Rivers, which are the greatest rivers of the Mediterranean in terms of freshwater fluxes, do not follow the general trend. Their outstanding behaviour may be explained by the non-Mediterranean climate in the upper northern parts of their basins.

Line 590: the role of dams. Why figure 8? Is it a claim of this model also to reproduce the monthly discharge? Why not show the whole timeserie? I would not include this figure.

In figure 8 we present a comparison of the seasonal cycle of monthly averaged water discharge between data and model outputs, because only monthly averaged data are available in [Vörösmarty et al., 1996] , i.e. the whole time series are not available from in-situ data. This figure illustrates the fact that seasonal variability (i.e. the succession of dry and wet periods) of water discharge is well simulated by the LPJmL-Med. However, as shown in Figure 8, the amplitude of the seasonal cycle is larger in the model because we do not represent explicitly the role of dams in the regulation of water flows (i.e. release and retention of water). Clarified in the revised ms.

Line 613: the word “could”? Are you not sure?

We agree that some aspects in the simulation still need to be improved, especially the inconsistency between model and in-situ data of NO3 for recent years: where data remained approximately constant, the model simulates à rapid increase of NO3 for almost all the rivers.

We removed this sentence in the revised manuscript.

Figure 9: There is something wrong with the model. The trend of observed NO3 is not reproduced by the model. This is a problem, because it is becoming worse when the time is closer to the current

situation. The same holds for Figure 10 for PO4. Same questions for these two figures? Where in the rivers are these comparisons?

The in-situ data used for comparison again come from the same locations as river gauging stations (see above).

For, figure 9 we agree with the referee concerning the modelled trend of NO3 (but not for PO4), which leads to an overestimation of NO3 in rivers by the model. We are aware that the model still needs to be improved (as mentioned in the submitted manuscript lines 717-721). We have worked on the representation of some of the involved processes in the model, especially the nitrification process, but the scarcity of NH4 data makes difficult the assessment of the model skill for this state variable).

However, the new simulation run for the revised version (due to the problem pointed by the referee concerning the NH4/NO3 ratio of sewage affluent, cf. answer above and Fig. R3) provides an improved representation of NO3 content in water discharge (cf. Figure R3). Moreover, it could be verified through very long simulations (RCP 8.5 regional climate scenarios) in the frame of an on-going work, that the slope of NO3 decreases rapidly after years 2000 and that NO3 provided by river outputs get stabilized.

In conclusions the word "concentration" is used. I did not see any concentrations in this article.

The LPJmL modelling community generally use the concept of content of the soil layers. Here as presented in the first part of section 2.2; we used the term of content (in g) for nutrients in soil, and concentration for those in water (in g.m^{-3}), with a daily time-step for all variables. However we converted the outputs of model to kt/y because all the available in-situ data are presented in this unit, and it is the unit generally used by the community.

References

[Billen and Garnier, 2007] Billen, G. and Garnier, J. (2007). River basin nutrient delivery to the coastal sea: Assessing its potential to sustain new production of non-siliceous algae. *Marine Chemistry*, 106:148–160.

[Bouwman et al., 2002] Bouwman, A. F., Boumans, L. J., and Batjes, N. H. (2002). Modeling global annual n2o and no emissions from fertilized fields. *Global Biogeochemical Cycles*, 16:28–1.

[Cerdan et al., 2010] Cerdan, O., Govers, G., Bissonnais, Y. L., Oost, K. V., Poesen, J., Saby, N., Gobin, A., Vacca, A., Quinton, J., Auerswald, K., Klik, A., Kwaad, F. J., Raclot, D., Ionita, I., Rejman, J., Rousseva, S., Muxart, T., Roxo, M. J., and Dostal, T. (2010). Rates and spatial variations of soil erosion in europe: A study based on erosion plot data. *Geomorphology*, 122:167–177.

[Desboeufs et al., 2018] Desboeufs, K., Nguyen, E. B., Chevaillier, S., Triquet, S., and Dulac, F. (2018). Fluxes and sources of nutrient and trace metal atmospheric deposition in the northwestern mediterranean. *Atmospheric Chemistry and Physics*, 18:14477–14492.

[ElElla, 1993] ElElla, A. (1993). Preliminary studies on the geochemistry of the Nile river basin, Egypt.

[Elser and Urabe, 1999] Elser, J. J. and Urabe, J. (1999). The stoichiometry of consumer-driven nutrient recycling: Theory, observations, and consequences. *Ecology*, 80:735.

[Gezerman, 2020] Gezerman, A. O. (2020). A novel industrial-scale strategy to prevent degradation and caking of ammonium nitrate. *Heliyon*, 6.

[Heffer, 2013] Heffer, P. (2013). Assessment of fertilizer use by crop at the globallevel 2010–2010/11. *International Fertilizer Industry Associ-ation, Paris.*

[IFA, 2016] IFA (2016). Ifadata.

[Isherwood, 2010] Isherwood, K. F. (2010). Fertilizer use in western europe: types and amounts, in: Interdisciplinary and sustainability issues in food and agriculture-volume iii.

[Longo et al., 2014] Longo, A. F., Ingall, E. D., Diaz, J. M., Oakes, M., King, L. E., Nenes, A., Mihalopoulos, N., Violaki, K., Avila, A., Benitez-Nelson, C. R., Brandes, J., McNulty, I., and Vine, D. J. (2014). P-nexfs analysis of aerosol phosphorus delivered to the mediterranean sea. *Geophysical Research Letters*, 41:4043–4049.

[Ludwig et al., 2009] Ludwig, W., Dumont, E., Meybeck, M., and Heussner, S. (2009). River discharges of water and nutrients to the mediterranean and black sea: Major drivers for ecosystem changes during past and future decades? *Progress in Oceanography*, 80:199–217.

[Mayorga et al., 2010] Mayorga, E., Seitzinger, S. P., Harrison, J. A., Dumont, E., Beusen, A. H., Bouwman, A. F., Fekete, B. M., Kroeze, C., and Drecht, G. V. (2010). Global nutrient export from watersheds 2 (news 2): Model development and implementation. *Environmental Modelling and Software*, 25:837–853.

[Mooshammer et al., 2014] Mooshammer, M., Wanek, W., Zechmeister-Boltenstern, S., and Richter, A. (2014). Stoichiometric imbalances between terrestrial decomposer communities and their resources: Mechanisms and implications of microbial adaptations to their resources.

[Morée et al., 2013] Morée, A. L., Beusen, A. H., Bouwman, A. F., and Willems, W. J. (2013). Exploring global nitrogen and phosphorus flows in urban wastes during the twentieth century. *Global Biogeochemical Cycles*, 27:836–846.

[Müller et al., 2012] Müller, J., Werner, K., Stein, R., Fahl, K., Moros, M., and Jansen, E. (2012). Holocene cooling culminates in sea ice oscillations in Fram Strait. *Quaternary Science Reviews*, 47:1–14.

[Neitsch et al., 2002] Neitsch, S. L., Arnold, J. G., Kiniry, J. R., and Williams, J. R. (2002). Soil and water assessment tool user's manual version 2000.

[Neitsch et al., 2005] Neitsch, S. L., Arnold, J. G., Kiniry, J. R., and Williams, J. R. (2005). Soil and water assessment tool theoretical documentation.

[Nixon, 2003] Nixon, S. W. (2003). Replacing the Nile: Are Anthropogenic Nutrients Providing the Fertility Once Brought to the Mediterranean by a Great River? *AMBIO: A Journal of the Human Environment*, 32(1):30–39.

[Pedde et al., 2017] Pedde, S., Kroeze, C., Mayorga, E., and Seitzinger, S. P. (2017). Modeling sources of nutrients in rivers draining into the bay of bengal—a scenario analysis. *Regional Environmental Change*, 17:2495–2506.

[Porqueddu et al., 2016] Porqueddu, C., Ates, S., Louhaichi, M., Kyriazopoulos, A. P., Moreno, G., del Pozo, A., Ovalle, C., Ewing, M. A., and Nichols, P. G. (2016). Grasslands in 'old world' and 'new world' mediterranean-climate zones: Past trends, current status and future research priorities. *Grass and Forage Science*, 71:1–35.

[Potter et al., 2011] Potter, P., Ramankutty, N., Bennett, E. M., and Donner, S. D. (2011). Global fertilizer and manure, version 1: Phosphorus in manure production.

[Raimbault and Lagadec, 2012] Raimbault and Lagadec (2012). Moose water sample analyses rhône river historical data.

[Reckling et al., 2014] Reckling, M., Preissel, S., ZALF, P. Z., Topp, K., Watson, C., Murphy-Bokern, D., and Stoddard, F. (2014). Legume futures report 1.6 effects of legume cropping on farming and food systems.

[Rost et al., 2008] Rost, S., Gerten, D., Bondeau, A., Lucht, W., Rohwer, J., and Schaphoff, S. (2008). Agricultural green and blue water consumption and its influence on the global water system. *Water Resources Research*, 44.

[Ryan et al., 2009] Ryan, J., Ibrikci, H., Sommer, R., and McNeill, A. (2009). Chapter 2 nitrogen in rainfed and irrigated cropping systems in the mediterranean region.

[Sattar et al., 2014] Sattar, M. A., Kroeze, C., and Strokal, M. (2014). The increasing impact of food production on nutrient export by rivers to the bay of bengal 1970-2050. *Marine Pollution Bulletin*, 80:168–178.

[Schaphoff et al., 2018] Schaphoff, S., Bloh, W. V., Rammig, A., Thonicke, K., Biemans, H., Forkel, M., Gerten, D., Heinke, J., Jägermeyr, J., Knauer, J., Langerwisch, F., Lucht, W., Müller, C., Rolinski, S., and Waha, K. (2018). Lpjml4-a dynamic global vegetation model with managed land-part 1: Model description. *Geosci. Model Dev*, 11:1343–1375.

[Skliris et al., 2007] Skliris, N., Sofianos, S., and Lascaratos, A. (2007). Hydrological changes in the mediterranean sea in relation to changes in the freshwater budget: A numerical modelling study. *Journal of Marine Systems*, 65:400–416.

[Stagnari et al., 2017] Stagnari, F., Maggio, A., Galieni, A., and Pisante, M. (2017). Multiple benefits of legumes for agriculture sustainability: an overview.

[Sterner and Elser, 2002] Sterner, R. W. and Elser, J. J. (2002). *Ecological stoichiometry: The biology of elements from molecules to the biosphere.* Princeton University Press.

[Strokal, 2013] Strokal, C. K. M. (2013). Nutrient export by rivers to coastal waters of china: An integrated modelling approach — research@wur. *Environmental Systems AnalysisWIMEK*.

[Thieu et al., 2009] Thieu, V., Billen, G., and Garnier, J. (2009). Nutrient transfer in three contrasting nw european watersheds: The seine, somme, and scheldt rivers. a comparative application of the seneque/riverstrahler model. *Water Research*, 43:1740–1754.

[Van Drecht et al., 2009] Van Drecht, G., Bouwman, A. F., Harrison, J., and Knoop, J. M. (2009). Global nitrogen and phosphate in urban wastewater for the period 1970 to 2050. *Global Biogeochemical Cycles*, 23(3).

[van Puijenbroek et al., 2018] van Puijenbroek, P. J., Beusen, A. H., and Bouwman, A. F. (2018). Datasets of the phosphorus content in laundry and dishwasher detergents. *Data in Brief*, 21:2284–2289.

[Von Bloh et al., 2018] Von Bloh, W., Schaphoff, S., Müller, C., Rolinski, S., Waha, K., and Zaehle, S. (2018). Implementing the nitrogen cycle into the dynamic global vegetation, hydrology, and crop growth model LPJmL (version 5.0). *Geoscientific Model Development*, 11(7):2789–2812.

[Vörösmarty et al., 1996] Vörösmarty, C. J., Fekete, B. M., and Tucker, B. A. (1996). Global River Discharge Database (RivDIS V1.0), International Hydrological Program. *Global Hydrological Archive and Analysis Systems, UNESCO, Paris*.

[Wang et al., 2019] Wang, M., Hu, C., Barnes, B. B., Mitchum, G., Lapointe, B., and Montoya, J. P. (2019). The great atlantic sargassum belt. *Science*, 364:83–87.

[Zessner et al., 2005] Zessner, M., Gabriel, O., Schilling, C., Lampert, C., Behrendt, H., Schreiber, H., Strauss, P., Laszlo, F., and Literathy, P. (2005). Outlines of a handbook for international nutrient balances on catchment scales including minimum-requirement-data-lis.