#### Authors' response to:

# Interactive comment on "Detection and attribution of global change effects on river nutrient dynamics in a large Mediterranean basin" by R. Aguilera et al.

#### Anonymous Referee #1

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The manuscript by Aguilera et al. deals with spatio-temporal patterns of nitrate and phosphate concentration in the Ebro river basin and tries to explain them by climate variability (seasonality, NAO) and anthropogenic impacts (land use, fertilizers, irrigation, river damming, waste water inputs). For that, they use 31yr timeseries data (1980-2011) from 50 sampling location distributed over the Ebro river and its tributaries. For 37 sampling locations, they additionally analyze time series of stream flow. They use dynamic factor analyses to extract common temporal patterns of all the time series. The identified seasonal cycles, multi-annual cycles, and long-term trends. However, these patterns have a different weight at each sampling location and a substantial proportion of sampling locations shows opposite trends. Then, the authors use multivariate statistics to analyze the relation between the factor loadings associated to each pattern at each sampling location and the differences in catchment properties and other environmental drivers. Overall, the manuscript is of potential interest for the readership of Biogeosciences. I would suggest the publication of the manuscript after some moderate revisions. In most of its parts, the manuscript is well written and the methods are clearly enough described. At some points, some clarifications are needed. In the following, I give some specific comments on methodology and results followed by more general comments on the text.

We thank Referee #1 for their constructive criticism and detailed comments and suggestions. We addressed these in the revised version of the manuscript and provide here the corresponding specific answers to each of the Referee's suggestions. The original Referee's comments are indicated in italics.

#### #1 In-stream/in-reservoir processes - catchment area

The authors analyze different potential drivers of the spatial-temporal patterns of fluvial nitrate and phosphate concentrations. Most of the identified drivers relate to the catchment properties and the sources of nitrate and phosphate. For the temporal patterns, in-stream and in-reservoir processes (in particular nutrient uptake by algae growth) play an important role as well. Consequently, the authors analyze reservoir capacity and location as potential environmental drivers. A potential driver of spatial differences in temporal patterns which could easily be addressed as well would be catchment area. This might not be a driver which changes over short time-spans, but, as a surrogate measure for average water traveling time, an important explanatory variable for the different identified patterns. The cluster analysis in section 4.3 and the related figures 3 and 4 suggest an upstream-downstream pattern and catchment area as explanatory variable seems thus promising. For instance, the pattern 3 identified for nitrates seem to become more important in upstream direction (clusters 4->1->2->3).As I get it from the methods section (section 3.7), land use (i.e. different nopoint sources of nutrients) is calculated once for the whole catchment and once for a buffer area around the sampling location. This provides the possibility to distinguish between non-point sources (i.e. agricultural areas) that are more upstream and those not far from the sampling location. For the latter, in-stream transformation and retention processes play less a role than for the nutrient loads coming from farther upstream, due to the shorter traveling time. The catchment area could maybe add valuable information. With increasing catchment area, on the one hand, the average traveling time of the water coming from upstream increases, and, on the other hand, the relative contribution from the 10 km buffer area decreases.

We agree that catchment area plays a role in shaping spatial differences in temporal patterns of nutrient concentration in river basins. For this reason, we originally included the total upstream catchment area in the land use explanatory variables. In other words, we used the areas of the upstream catchments to specific monitoring points for each of the land uses we considered; Industrial, Urban, Dryland and Irrigated agriculture, Forest, Grassland, and Water, all of which were expressed in km<sup>2</sup>. In addition, in order to depict more local conditions, these land uses were also included as explanatory variables with values obtained within a 10 km radius of each sampling point.

Additionally, meteorological variables such as precipitation and air temperature, as well as water and land management variables such as reservoir capacity and fertilizer application were also introduced as the sum of the values in upstream catchments to each specific sampling point, reflecting in this way the catchment area factor.

The use of catchment area as a separate explanatory variable added a high degree of collinearity with the relevant above mentioned variables. Catchment area was therefore excluded in our analysis. This is now specified in the Methods section in Page 9 of the revised manuscript.

In section 3.7, the authors write that they consider "reservoir capacity and location, waste water treatment plants (WWTP) discharge and location". From the manuscript, it does not get clear to me how they consider the location of reservoirs and WWTPs. This would be important to know, because the location (immediately upstream or farther upstream?) would likely have an effect.

We considered the total capacity of reservoirs and the total discharge from upstream catchments for each sampling point as the farther upstream component. By separately considering the capacity and discharge of these explanatory variables (reservoirs and WWTP) immediately upstream the sampling point within the 10km buffer, we differentiated between more local effects and regional effects. An additional sentence was introduced in this Section to clarify this point (Page 9, Lines220-222 of the revised manuscript).

## #2 Instream/reservoir processes part 2 N vs P

The spatio-temporal patterns of nitrate and phosphate concentrations might influence each other. With regard to algae uptake of nitrate and phosphate, it would be interesting to know what is the limiting factor of algae growth in the basin. Is it either nitrate or phosphate, or another factor (like light limitation)? Nitrate and phosphate show different long-term trends, with phosphate decreasing in the 1990s and nitrate somewhat later (large rivers). Does this have an effect on in-stream/reservoir algae growth and nutrient uptake/retention?

A very significant decrease in the concentration of the dissolved phosphorus was observed after the mid 90s. This decrease coincides with the improvement of urban sewage treatment in the most important cities of the Ebro basin. According to a study carried out by Ibañez et al., (2008) there was a significant positive correlation between the concentration of dissolved phosphorus and the concentration of total chlorophyll between the 1987-2004 period (Page 19 of the revised manuscript). Here, low flow conditions together with decreasing dissolved phosphorous and decreasing phytoplankton were likely the main factors causing the increase of water transparency, which improved the eutrophy condition. The results of this study suggest that the observed changes in chlorophyll (first increasing and then decreasing) in the lower Ebro River were a direct consequence of the changes in phosphorus and the DIN/DIP ratio.

## #3 Land use change

The authors analyze time-series of nitrate and phosphate over the 31 yr period 1980 to 2011. They explain differences in increasing and decreasing trends by the areal proportion of different land use types. What time is represented by the used land use data? Was there a significant change in land use in the Ebro Basin over the last three decades?

Re-vegetation is the most significant catchment change that has occurred in the mountainous areas of the Ebro basin during the 20th century. The onset of farmland abandonment and re-vegetation is set between 1950 and 1960 (Garcia-Ruiz et al., 1996). The land use conditions included in our study represent the average conditions between the period 1980 to 2011,

where no other significant or drastic land use changes occur, other that management practices related to the improvement of industrial and urban wastewater, which is reflected in the decrease of phosphate in the mid 1990s. The latter sentence was included Section 3.7 for further clarification (Page 9, Lines 213-216 of the revised manuscript).

#### #4 Climate change

The authors argue that climate change would have an effect on nutrient and phosphate concentration. They identify, however, only effects of climate on the seasonality and multi-annual cycles of phosphate and nitrate concentration which could be related to climatic seasons and the NAO. To show the effect of CLIMATE CHANGE on nitrate and phosphate concentration, they would need to identify a correlation between long-term trends in nutrient concentrations and climatic variables.

We wanted to reflect the effect of climatic variables in the spatio-temporal distribution of dissolved nutrients in the Ebro basin. We found that streamflow and air temperature shaped nitrate patterns, and that regional and global climatic modes influenced the variability of nutrients at the basin scale. In a sense, the changes in these climatic modes within the 31 years included in our study could indicate the potential role of climate change in in-stream nutrient variability. Regarding long term trends, we did not find a significant correlation between nutrients and climatic variables. This is now discussed in Section 5.1 of the revised manuscript (Pages 17-18)

#### #5 Nutrient fluxes from land to sea

The lateral fluxes of nitrate and phosphate would be more interesting than the concentrations, because they directly describe the inputs of nutrients to the river or the exports of nutrients to the coast. The fluxes could be easily compared if they were reported relative to the catchment area (e.g. t N km-2yr-1 or moles m-2yr-1). It would be interesting for the readers what the spatio-temporal patterns of nutrient fluxes would be. Also for the long-term trend it would be more interesting to see if the flux of nutrients increased/decreased, in particular for the sampling location which is farthest downstream (because this sums up all the changes upstream and represents the final export to the coast).

We mainly dealt with nutrient concentration as we wanted to study the temporal and spatial distribution of nutrients in the river network of the Ebro basin and to relate these in-stream concentrations to potential sources of impact related to global change phenomena. Nevertheless, exploring the fluxes that ultimately reach coastal waters is also interesting. For this reason, and as suggested in this comment, we have included the fluxes in two key stations: Downstream-Tortosa and Upstream-Mendavia, both on the Ebro River (Supplementary Material in the revised manuscript). We however did not perform DFA for these fluxes due to computational difficulties related to the complexity of these analyses and the time restrictions for the revision of this manuscript.

Regarding the long-term trends, the overall decrease of phosphate flux is reflected in both upstream and downstream sampling points shown in the Supplementary Material (Section S.1). Significant long-term trends in nitrate flux were not identified.

**#General Comments** <u>Introduction</u> Page 5261 L 6: You should try to find a more suitable word for "action". Maybe "impact"?

The word action is in the original definition of global change by the US Global Change Research Act, we therefore maintain this word in this particular line. Impacts of global change

phenomena on freshwater resources are mentioned in the following lines (Page 2 in the revised manuscript).

# L12-15: Please, shortly explain here why this would be a fundamental concern.

Nutrient pollution derived from anthropogenic activities impacts inland and coastal waters, resulting in serious environmental and human health issues, and impacting the economy. A brief referenced explanation has been added to the text (Page 2, Lines 40-42 in the revised manuscript).

L22-24: I don't really understand this sentence. Are you talking about the eutrophication of the rivers themselves (then the concentrations of nitrate and phosphate in the water would be important) or about the eutrophication of the coastal waters (then the fluvial nitrate and phosphate fluxes would be important).

In this context, we are mainly referring ot the eutrophication of rivers and inland waters themselves, which is also why we work with nutrient concentration values instead of fluxes. The sentence has been rewritten (Page 2 in the revised manuscript).

#### Page 5262

L1-3: Do you really mean "insight of the physical, biological, or socioeconomical events"? Or rather the impacts of these events?

By extracting the key properties of time-series one can obtain evidence of changes and hints of potential causes behind such changes, which are later corroborated with comprehensive analyses. In a sense, one thus obtains information about the potential events that might have caused the observed impacts on the time-series being analyzed. This sentence has been nonetheless slightly modified to clarify our point (Page 3 in the revised manuscript).

## L13-19: Maybe you should shortly explain and evaluate (strengths, shortcomings) of all of each methods.

Although the evaluation of these methods is out of the scope of this paper, we emphasize some strengths and shortcomings of the different methods in the following lines (Page 3, Lines 75-83 in the revised manuscript), such as the inability of extracting common patterns from sets of time-series and not being able to deal with missing observations.

# L20: "Spectral analysis" was not mentioned before. What do you mean by "methods like spectral analyses"? Does this include all the methods named above?

We meant that spectral analysis methods, such as Singular Spectral Analysis, as well as the previously mentioned methods related to trend analysis and time-series analysis in general, are not able to simultaneously extract common patterns from a set of time-series. The sentence in the previous manuscript has been modified to exemplify spectral analysis methods (Page 3 in the revised manuscript).

L24-28: The meaning of this sentence is not clear to me. As I get it from the text, you need a good data coverage to identify local stressors and disentangle their effect from the effects of global stressors. Thus, you try to avoid discarding time-series from your data pool and rather opt for an advanced method which can get valuable information out of less-consistent time series. If that is the case, you should clarify this here and write it in a more comprehensible way.

Yes, we wanted to avoid discarding time-series in our dataset and therefore chose a method that could simultaneously deal with sets of time-series and cope with data gaps. The sentence has been modified to emphasize this idea (Page 4 in the revised manuscript).

# <u>3 Methods</u>

Page 5264

L14-15: How do you defined patterns? Are these the temporal patterns, i.e. seasonality, long-term trends and multi-annual cycles? Please, clarify here.

We make reference to the temporal patterns, such as cycles and trends. This line has been modified to make this clearer (Page 5 in the revised manuscript).

*L16: The abbreviation 'DFA' should be defined. It appears here for the first time.* 

The abbreviation and its definition first appears on Page 5263 (Introduction), and are also included in the abstract. We however have defined DFA also in this line (Page 5 in the revised manuscript).

Page 5266

L12-13: "significant trends that are not necessarily a straight line". Better use formulations like "non-linear trend".

The line has been changed to the suggested formulation (Page 7 in the revised manuscript).

Page 5268 L3-4: Do you have a reference for this?

A reference has been included to support the idea that generalized least squares for regression modeling is advisable when neighboring values of the response variable tend to be spatially correlated (Page 9 in the revised manuscript).

L5-8: What is a "spatial error structure"? What are the other 5 options for error structures? Why is the Gaussian structure (=" Gaussian distribution" ?) the best option?

This sentence has been modified to clarify the implementation of the Gaussian distribution as the spatial error structure, which was the best option for our generalized least squares (gls) models fitted by means of the nlme R-Package (Pinheiro et al., 2012) (Page 9 in the revised manuscript).

# <u>4 Results</u>

Page 5270

L1-3: How significant is that trend, when 20 of the 50 stations show an opposite trend? Also in Fig. 1c, this trend is not visible.

The trend is not visible as nitrate pattern 3 was not the dominant pattern (i.e., it had a negligible factor loading magnitude) in this particular sampling point (Miranda de Ebro), located in the upstream section of the basin. The significance or relevance of this opposite decreasing trend in nitrate concentration is indicated by the magnitude of the factor loadings in those 20 stations, shown in Figure 2.

In Table 2, the authors list the identified potential drivers of all identified patterns, also for pattern 3. For pattern three, they make the distinction between stations with a positive factor loading and stations with a negative factor loading. Interestingly, for both they identified 'Industrial area (%) UPSTREAM' as explanatory variable with the same positive coefficient. What does that mean? Please, discuss.

The role of the Industrial area (%) UPSTREAM explanatory variable and the same sign for nitrate Pattern 3 with positive factor loadings (decreasing long-term trend) and with negative factor loadings (increasing ling-term trend) could be explained by the fact that Pattern 3 was particularly relevant (i.e., factor loading magnitudes were higher, regardless of their sign) in areas with little industrial activity. What made the difference between the decreasing versus the increasing trends, in addition to the other significant explanatory variables identified, could have been the varying types of industrial activities present in the vicinities of particular sampling points. The information on specific types and impacts of industrial activities in the basin was not available.

#### Page 5271

L3-17: Here, it would be interesting to see a similar pattern analysis for stream flow, because the authors identified a clear relation of nitrate pattern 1 to stream flow (Fig. 1e). Next, it would be interesting to see if stations with different factor for nitrate pattern 1 would also show different factor loadings for any identified pattern of stream flow.

We extracted common patterns from streamflow time-series in 37 sampling points in the basin. The relevant results related to the coherent cycles of streamflow with nutrient concentration and climatic variables are presented in the paper. However, carrying out a full analysis of the DFA streamflow patters, including all the steps outlined in the Methods section, would have considerably extended the length of the manuscript and potentially hindered the interpretability and the main scope of this paper. For this reason, these more specific analyses are not included here.

Also, there was no factor loading sign switching among the extracted patterns for streamflow.

Table 2: For nitrate pattern 1 – positive factor loadings, Mean air temperature (upstream) was identified as an important explanatory variable. This variable was also identified to show a strong negative correlation to nitrate pattern two (Fig. 1e). In Fig 1e,f, the nitrate patterns 1 and 2 do not seem that different, with a minimum in late summer, when average air temperature is highest. This is an issue that would have to be discussed.

The significance of mean air temperature as an important explanatory variable for nitrate pattern 1 (positive factor loadings) has to do with the spatial distribution of the mean temperature values and the (negative) relationship between these two, identified by means of gls regression models.

The relationship between temperature and Pattern 2 in Fig 1e. is based on averaged temperature values for the Ebro basin.

Furthermore, although there is a common minimum in late summer between nitrate patterns 1 and 2, there two patterns are overall very different from each other and were clearly and significantly related to two different variables, as shown in Table 2 and Figures 1e and 1f.

Page 5272

L19-21: The authors showed that nitrate pattern 2 can be correlated to temperature and this might be due to biological activity, or phenology like the authors expressed it. While reading 'phenology', I think about terrestrial vegetation and, in this context here, the control of terrestrial nutrient cycling on the exports of

nutrients to streams. Here, the authors show that this pattern (pattern two) is most dominant in the far downstream part of the Ebro. If a terrestrial control was the cause of this temporal pattern, it would be interesting to know why this pattern is less dominant in more upstream parts. Might it be that this pattern is due to in-stream uptake of N and P by aquatic autotrophic production? Then, 'phenology' might be a bit misleading.

As stated in the case of nitrate concentration, assimilation by freshwater primary producers during summer and the seasonal evolution of leaf fall and decomposition could have taken a major role. These factors are grouped in the term phenology, which is not restricted to terrestrial ecosystems, but can also include the activity of freshwater algae.

Nevertheless we have stated that the nitrate pattern 2 related to temperature in the Ebro basin gained more relevance in the downstream rivers and streams due to the presence and control of large reservoirs and the biogeochemical processes occurring therein and immediately downstream (Page 14 in the revised manuscript).

We include a discussion of the potential downstream shift from terrestrial phenology to biogeochemical reservoir processes as biological control of pattern 2 in the Discussion section of the revised manuscript (Page 15), as suggested by the Referee in the following comment.

## 5 Discussion

Page 5273, L21-Page5274, L4: See comment before. The nitrate pattern 2 seems to be most dominant in the downstream part. Could this indicate that algae growth has a more dominant effect than phenology of terrestrial ecosystems? Or is this due to the fact that pattern 1 is lower (due to retention in reservoirs) and thus the relative contribution of pattern 2 is higher? But then, the pattern 2 driven by terrestrial ecosystem phenology would also be attenuated due to water retention in reservoirs and, thus, algae growth would be left as the driver for pattern 2 in the downstream section of the Ebro. Maybe you should discuss the potential downstream shift from terrestrial phenology to algae growth as biological control of pattern 2.

Terrestrial phenological processes such as those involved in leaf fall and decomposition would potentially be more important in upstream sections of the basin, where the biogeochemical activity in large reservoirs is not present. Based on this and the previous Referee's comment, we have specified the effect of downstream reservoir biogeochemical control on nitrate pattern 2 in the Discussion of the revised manuscript (Page 15).

Page 5275: If the authors also showed their results from the DFA for stream flow, like they did for nitrate and phosphate in Fig 1, this could help interpreting and discussing the effects of NAO and ENSO on the patterns of nitrate and phosphate. So far, from figure 1, only the average 12 month cycle of stream flow is visible. In table 1, they also state stream flow oscillations at 1.5, 2.2, 3.2, and 4.2 yrs. It would be interesting to have these identified patterns for stream flow as a plot which could be directly compared to those for phosphate and nitrate. It would also be interesting to see if there is a longterm trend for stream flow, in particular at the site farthest downstream.

As stated earlier in this document, we extracted common patterns from streamflow timeseries in 37 sampling points in the basin. Adding all this information in the manuscript would detriment its current flow and its main scope. For this reason, we have added the DFA results for streamflow in Section S.2 of the Supplementary Material.

No significant correlation was found between long-term trends for streamflow and nutrients in the Ebro basin, and this is indicated in Table 1 for each pattern with a significant long-term trend identified by the Kendall tau and p-values in the yue-Pilon trend analyses. In fact, we did not identify any significant trend in streamflow common patterns identified by means of DFA.

Section 5.2, first paragraph: Here, I got a bit confused and had to read through the text several times. From Page 5272, L27 to Page 5276, L4: Do you refer to the sampling locations with increasing trends? If yes, please clarify that in the text.

We refer to all significant trends identified for both nitrate and phosphate; we have clarified this idea in the revised manuscript.

You should try to restructure the whole paragraph and make it more logical. For the explanation of decreasing vs. increasing trends, you should start with the terrestrial sources: what human activities might have decreased nitrate concentrations (e.g. more rational application of synthetic fertilizers, improved sewage water treatment) and what might have increased nitrate concentrations at other sampling locations. Then you should come to the differences related to upstream-downstream patterns. Of course, head water streams might show stronger increasing trends if the sources increased, and decreasing trends when the sources decreased. Smaller catchments are likely more homogenous than larger catchments, that means that it is more likely that either decreasing or increasing terrestrial inputs prevail. Larger catchments, in particular because the catchments here are nested and large catchment contain multiple small sub-catchments considered here, will more likely contain a mix of increasing and decreasing terrestrial sources. Further, due to longer traveling times of the water, and additionally the impact of reservoirs, increasing nitrate inputs might also cause increased algae uptake (and denitrification?) that might attenuate increasing trends at downstream locations.

We have re-structured the first paragraph in Section 5.2 in the revised manuscript to include the suggestions stated above.