Dear editor,

Please find enclosed the revised version of the manuscript entitled "Reduced phosphorus loads from the Loire and Vilaine Rivers were accompanied by increasing eutrophication in Vilaine Bay (South Brittany, France)".

We thank you again for giving us the opportunity to revise our manuscript and are grateful to reviewers for their comments and suggestions on the earlier version. Following their suggestions, we have revised the manuscript and made substantial modifications.

In the **introduction**, the objectives became an investigation into long-term evolution in eutrophication parameters in the Loire and Vilaine Rivers and the Vilaine Bay (VB), with a working hypothesis that eutrophication trajectories in coastal waters in recent decades have been influenced by those in rivers. The study also aimed to establish the link between fresh and marine water trajectories and highlight the impact of N versus P reduction strategies in rivers on coastal water quality.

In the **materials and methods**, we provided elements showing the representativeness of the VB monitoring station (Ouest Loscolo). We added the contribution of Loire DIN inputs to VB DIN concentrations estimated by ECO-MARS3D model (Ménesguen et al., 2019), to establish better the links between the Loire and the VB. We also improved the description of tools used to assess the long-term trends and changes in seasonality by explaining more clearly the use of the monotonic Mann-Kendall test to corroborate the DLM results.

The **results** have been rewritten and reorganized and the DLM figures modified by transforming the *y-axis* into logarithmic scale so that the reader can access the actual values. We decided to remove some results: diatom/dinoflagellate ratios (Fig. 7g, h).

The **discussion** has been reorganized and extended:

1) Eutrophication trajectories at river basin outlet,
2) Eutrophication trajectories in the VB, including a section on the respective contributions of diatoms and dinoflagellates to eutrophication processes in the VB,
3) River – VB continuum, adding a section dedicated to the potential influence of the processes on nutrient loads within estuaries and dam,
4) Implications for nutrient management.

The comparison between benthic flux measurements and calculated riverine nutrient loads has been removed (Table 4)

We sincerely thank you and look forward to your positive feedback.

On behalf of the co-authors,

Widya Ratmaya

Referee 1 – C. Minaudo

General comments

Referee's comments (RC) - This study by Ratmaya et al., focuses on eutrophication trajectories over three decades in a large bay located in the French Atlantic coastal zone. It tries to link long term trends and seasonal evolutions in the main bay tributaries with the ones observed in the bay itself, based on a trend+seasonalilty time series decomposition algorithm. This study could be of interest for Biogeosciences readers, but suffers from too many issues such as lack of a clear research question, lack of structure within and between sections, and several technical issues that need to be addressed before it can be considered for publication.

The main issues to me are the following:

- Concentration time series in the Loire River (the main tributary) originate from a station located in a river section under estuarine influence but was considered as representative of the freshwater part.
- Methodology is not clear, especially for the seasonal analysis using the DLM approach. Authors need to define clearly the metrics that were used in this work (e.g. little is said on MK slopes p-values although they appear in Tables)
- Nothing is presented on the impact of estuarine zones on DIN and NIP, disabling the credibility of the interpretations made to explain eutrophication trajectory in the coastal zone.
- If the presence of a dam at the outlet of one of the two tributaries is mentioned, nothing is explained on the potential impacts this should have on the nutrient dynamics discharged into the bay Additionally, this manuscript needs language editing. Many sentences need to be either removed or modified for the sake of clarity. I decided to focus on specific comments on Method and Results sections, because I think interpretation in the Discussion section might change once everything has been addressed properly.

Author's comments (AC) - We thank the referee for the detailed and constructive comments. All issues raised are listed and carefully answered point by point below. The previous manuscript was carefully reviewed by an English native. However, the sentences that referee pointed out will be reviewed and modified if necessary.

Specific comments

1. **RC** - Page 2; Lines 28-29 (2;28-29): this hypothesis has been proven wrong in many studies. I don't think you should present your problematic this way.

   AC - The hypothesis tested in the present study deals with coastal waters, based on Schindler et al. (2008) and Schindler (2012), who stated that the reduction of P inputs is enough to mitigate eutrophication in lakes and other freshwater ecosystems. Although authors carried out their experiments on lakes, they wondered whether the P-only reduction paradigm could be applied to coastal waters. Schindler (2012) also stated that he was unable to find long-term, ecosystem-scale evidence that controlling N input, either alone or in addition to P resulted in oligotrophication of estuaries. We believe that our dataset provides the opportunity to demonstrate that, conversely, without N input reduction in rivers, the coastal waters under their influence are unlikely to recover from eutrophication.

Author's changes in manuscript:

We plan to review the introduction by integrating recent articles by Schindler et al. (2016), Paerl et al. (2016) and others (see letter to editor).

2. **RC** - Page 3, section 2.2: explain that you extracted the longest records available. The reader doesn't know at this point that multi-decadal data is available.

Author's changes in manuscript:

The information on the availability of dataset will be added to the main text of the manuscript.

3. **RC** - Page 3, section 2.2: If Montjean is considered as the last freshwater station on the Loire, why would you use concentrations originating from Ste Luce located in the zone influenced by estuarine salinity? This is a choice that could mislead your interpretations. Also, when computing loads, which site served as the reference? That means did you calculate loads at Montjean or Ste Luce and how did you proceed (e.g. catchment areas ratio?)?

AC - Sainte-Luce is the last station for water quality monitoring on the Loire, located upstream of the haline intrusion (Guillaud et al., 2008), therefore it is a freshwater station closer to the river mouth than Montjean. The influence of tidal dynamics at Sainte-Luce was avoided by discarding data collected during high tide. In the database of Loire-Brittany River Basin Authority, Sainte-Luce displays a longer dataset (since 1980s) than Montjean (from 1995). Nutrient concentrations measured at Montjean showed parallel long-term evolutions to those observed at Sainte-Luce (Figure R1).

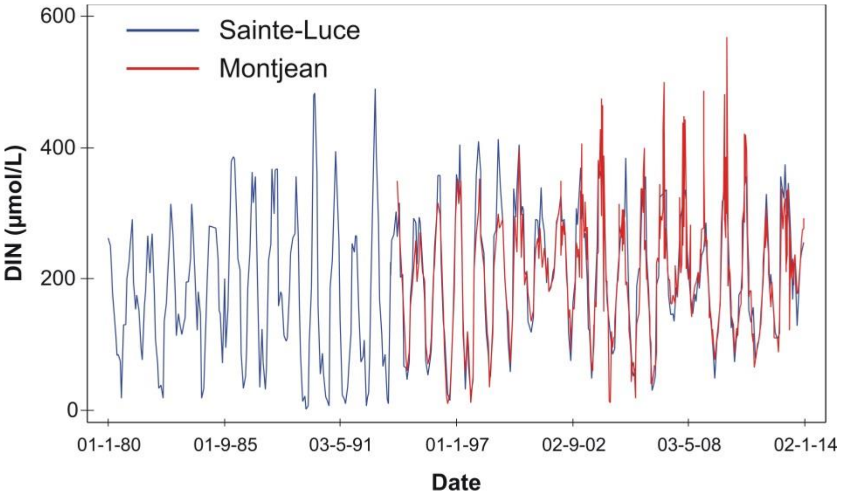

Figure R1. DIN concentrations in the Loire River at Sainte-Luce (blue) and Montjean (red)

Riverine nutrient load calculations were based on nutrient concentrations at Sainte-Luce and river discharge at Montjean, since there is no measurement of discharge at Sainte-Luce. Guillaud et al. (2008) calculated riverine nutrients loads based on the same stations, as a forcing parameter for the ecological ECO-MARS3D model simulating phytoplankton production in the Bay of Biscay (see Huret et al., 2013; Ménesguen et al., 2014; Ménesguen and Dussauze, 2015; Ménesguen et al., 2018).

Riverine nutrient loads were calculated as a function of river discharge and nutrient concentrations, not in relation to the catchment area (see below).

For this section, we decide to keep Saint-Luce as reference station for water quality. Exchanging Sainte-Luce dataset for that of Montjean will not affect the overall results.

Author's changes in manuscript:

"DIP, DIN and Chl a data came from stations located upstream of the haline intrusion: Sainte-Luce-sur-Loire on the Loire and Rieux on the Vilaine, and DSi from Montjean-sur-Loire on the Loire and Férel on the Vilaine (Fig. 1)."

The following lines will be added to the manuscript in order to clarify the choices of stations:

"For Sainte-Luce-sur-Loire, the influence of tidal dynamics was avoided by discarding data collected during high tide."

"In order to calculate riverine nutrient loads, gauging stations located close to the river mouth were selected and specifically those where nutrient concentrations were regularly measured. When information on nutrient concentrations was not available for the same location, data from the nearest station representative of the river outlet was used."

4. **RC** - Page 4, Line 4-5 (4;4-5): you should make sure this assumption on NO3 being >90% TN is correct. For the riverine part, AELB also provides TN concentrations.

AC - The referee seems to confuse dissolved inorganic nitrogen (DIN) with TN. In the present study, we considered dissolved inorganic nutrients (as bioavailable forms of nutrient for phytoplankton). Concerning DIN, nitrate was the most dominant form (>90%) of DIN (see Bouraoui and Grizzetti, 2011; Garnier et al., 2018; Ménesguen et al., 2018). Thus, our sentence was correct. However, we can improve the clarity of the sentence.

Author's changes in manuscript:

"DIN was defined as the sum of nitrate, nitrite and ammonium, with nitrate as the major component (>90%)."

5. **RC** - 4;6: this method for load calculations is subject to large errors, especially on DIP. You should use a discharge weighted method, commonly used by our community, and recommended within OSPAR convention.

AC - The method recommended by OSPAR, discharge weighted concentration (DWC) is commonly used when calculating annual loads, as also stated in Dupas et al. (2018) and in RID document (OSPAR Commission, 2017). This method is not relevant when long-term trend study includes seasonal variation. Moreover, this method has disadvantages. One of which is the application of limit of quantification when data is missing or unavailable. This can overestimate load estimation. Therefore, we prefer to retain our method of load calculation.

6. **RC** - 4;24: residuals as white noise is an hypothesis that is not always met by these algorithms. Please, remove "white noise" in this sentence.

Author's changes in manuscript:

"white noise" will be removed.

7. RC - We need a metric to assess if your algorithm performs well or note, especially when working at the seasonal scale with variables that don’t have stable seasonality patterns (e.g. phytoplankton biomass).

AC - In the present study, we use DLM to analyze time series data of water quality parameters linked to eutrophication. The DLM time-series analysis provides figures allowing the visual identification of changes in trends and in seasonality.

The trend plot displayed observed values with colored dots corresponding to the season. The trend was represented by a dark grey line with its 90% confidence interval (shaded area). For period 1997-2013, the longest common record for all variables, a linear trend significance test was performed on trend components from DLM using a modified non-parametric Mann-Kendall (MK) test (Yue and Wang, 2004). When linear trends were significant ($p<0.05$), their magnitude was calculated using non-parametric Sen’s slope estimator (Helsel and Hirsch, 2002).

The DLM seasonality plot displays the seasonal component estimated by the DLM, which indicates the seasonal position of maximum and minimum values, and the amplitude of seasonal cycles. Changes in the timing of annual maximum or minimum values were highlighted in the seasonality plots by colored dots, which change over time. Changes in the seasonal amplitude (increase or decrease of the value for a given season) were assessed using the modified MK test performed on DLM seasonal components from each season. The seasons were defined as: winter (January, February, March), spring (April, May, June), summer (July, August, September), and autumn (October, November, December).

Thus, the significance test of linear changes in DLM trends and seasonality components were provided by the modified MK test associated with Sen’s slope estimator.

Table R1 below shows the coefficient of determination for each model parameter, which gives an indication of the goodness of fit of a model. It is estimated by calculating the square of the sample correlation coefficient between the observed outcomes and the observed predictor values. It can also be viewed as the ratio of the explained variance to the total variance. We also add the estimated significance of trends and seasonality based on the squared correlation coefficient between the calculated trend and deseasonalized data and on the squared correlation coefficient between the calculated seasonal component and detrended data respectively (Minaudo et al., 2015).

Table R1. Coefficient of determination, significance of trends and seasonality estimated for the period of 1997-2013

Parameters	**Overall (%)**	**Trend (%)**	**Seasonality (%)**
Loire discharge	94.6	52.8	94.1
Loire DIP	51.5	16.9	45.6
Loire DIP Loads	71.3	29.4	67.1
Loire DIN	87.9	38.9	87.1
Loire DIN Loads	92.6	60.7	91.8
Loire DIN/DIP	48.1	18.6	41.6
Loire Chl a	68.8	55.6	53.3
Vilaine discharge	96.1	65.9	95.7
Vilaine DIP	52.0	40.0	28.4
Vilaine DIP loads	71.5	48.4	60.1
Vilaine DIN	95.7	60.1	95.4
Vilaine DIN loads	95.2	62.4	94.7
Vilaine Chl a	59.0	44.4	43.3
Vilaine DIN/DIP	66.3	42.3	55.5
VB DIP	61.0	4.9	60.5
VB DIN	85.7	33.4	85.0
VB DSi	64.3	9.9	62.6
VB DIN/DIP	83.2	23.5	82.7
VB DIN/DSi	78.9	18.0	78.1
VB DSi/DIP	28.1	16.9	15.6
VB Chl a	58.8	26.7	51.0
VB Diatoms	48.5	6.9	46.7
VB Dinoflagellates	43.7	2.7	43.0

8. **RC** - 5;1: why was this log-transformation necessary? It needs justification.

AC - The log transformation was necessary because:

“many measurements show a more or less skewed distribution. Skewed distributions are particularly common when mean values are low, variances large, and values cannot be negative, as is the case, for example, with species abundance, lengths of latent periods of infectious diseases, and distribution of mineral resources in the Earth's crust. Such skewed distributions often closely fit the log-normal distribution (Aitchison and Brown 1957, Crow and Shimizu 1988, Lee 1992, Johnson et al. 1994, Sachs 1997)."

This subject has been deeply discussed in Limpert et al. (2001).

Log-normal distribution induces a variance to mean relationships, that is, as in our case the mean and the variance vary with time, and thus the homoscedastic hypotheses, i.e., specifically for us, the equality of error terms variance through time, may not be fulfilled. This is why "A variance stabilizing log transformation…" is applied in the first place.

In addition, since all of our variables are positive, treating them without log-transformation may lead confidence intervals to include negative values, which consequently leads to inadequate models.

Author's changes in manuscript:

Justification of this log-transformation will be added in the revised manuscript.

9. **RC** - 5;7: reading log-transformed units is not convenient for the reader. You can log-transform the axis but still present actual values. Why did you log-transform the data in the first place? It makes the trend observation less clear to the reader.

Author's changes in manuscript:

We suggest log-transforming the axis to present the actual values. Please see an example Figure R2.

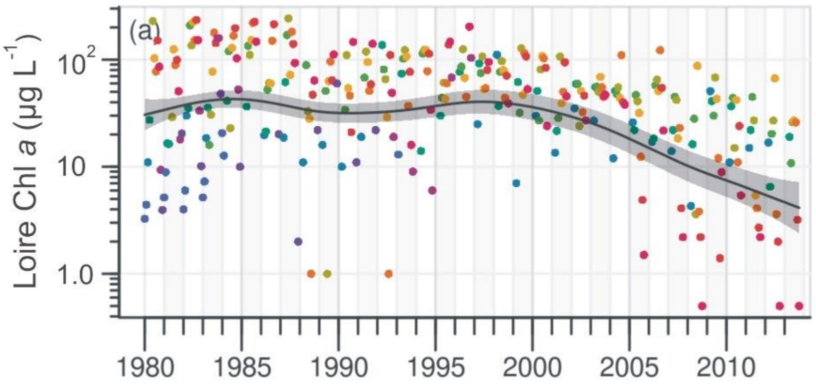

Figure R2. Trends of Chl a concentrations in the Loire

10. **RC** - The authors decided to use units that are consistent in the manuscript, but not commonly used by researchers on lotic environment. Please, convert all mol/L into mg/L or µg/L

AC - The SI unit of concentration (quantity of substance) is the mole per cubic meter (mol m^{-3}), which is commonly used in marine environment research. The use of "mol" is also consistent with the calculation of nutrient molar ratios, which permit us to assess potential nutrient limitation. Therefore, we prefer to keep as it is.

11. **RC** - 5;8-9: the explanation on trends significativity test is not clear nor properly justified. You need a metric for this. Why not use Sen's Slope significativity test?

AC - Please see the detail for trend significance test in point #7.

Author's changes in manuscript:

The use of the modified Mann-Kendall test as a trend significance test will be more detailed in the section 2.4.

12. **RC** - 5;11: the authors should define clearly which metrics were extracted for the time-series analysis and used for further analysis.

    AC - Please see point #7 for trend significance test.

13. **RC** - 5;15: have you conducted MK test on de-seasonalized = observations – seasonal component, or on de-seasonalized = trend component? The latter discards residuals from the analysis and this choice should be justified. Also, I think residuals from your DLM algorithm should be plotted along.

    AC - The modified Mann-Kendall test was conducted on de-seasonalized = trend component from DLM (please see point #7 for details).

    We provide residual plots for all parameters treated in this study in Appendix 1 for the consideration and verification of the referee. We did not include residual plots in results, as the number of figures was already high, as noted by the referee in point #21. Furthermore, papers including such figures are very uncommon.

14. **RC** - 5;17: it is not clear how you proceeded to identify seasonal trends. Did you use a seasonal MK test? This needs more details since it is the core of your analysis.

    AC - Please see point #7 for detail in seasonal trends.

15. **RC** - Besides, how would you justify analyzing loads evolutions and not only concentrations since you show that Q was stable over time? Removing all the load trajectory description would save space for other elements in your paper, and benefit to the clarity of your messages.

    AC - River discharge (Q) appeared stable in spite of oscillations. The modified MK test applied to river discharge trend component from DLM showed a significant decrease between 1997 and 2013 (*p*<0.05, Table 1). Therefore, it was necessary to calculate riverine nutrient loads in order to show that these loads displayed similar trends to those of nutrient concentrations.

16. **RC** - 5;21: What is STATGRAPHIC CENTURION and what are the metrics/analysis conducted with this? Please, add a reference for this.

    AC - In the manuscript section 2.4, we mentioned the use of STATGRAPHIC CENTURION software for Spearman Correlation analysis.

    Author's changes in manuscript:

    We suggest a modification if needed as follows:

    "Spearman Correlations were computed for annual median values in order to analyze relationships among variables, and tested using STATGRAPHIC CENTURION software (Statgraphics Technologies Inc, Version XVII, Released 2014)."

17. **RC** - 5;26: how significant is this trend in Q data? A large slope in MK tests doesn't mean that it is statistically significant.

    AC - Table 1 shows a *p*-value of 0.014 (for significance level of 0.05) for river discharge, indicating a significant trend. The negative slope indicates a decreasing trend. This decrease in Loire discharge was also observed in previous studies (please see point #15)

18. **RC** - 6;5: this seasonal shift is not observable in Figure 3. Consider adding a Figure to show seasonal variations and evolutions.

AC - The seasonal shift in position of annual DIP minimum from summer to spring is clearly visible by the change in color (Fig. 3b). It started with yellow (summer) around 1999 and changed to green (spring) from 2007 to the end of studied period.

Author's changes in manuscript:

We modified sentences concerning the seasonality as follows.

“Changes in position of annual maximum or minimum were highlighted in the seasonality plots by colored dots, which change over time. Changes in the seasonal amplitude (increase or decrease of the value for a given season) were assessed using the modified MK test performed on DLM seasonal components from each season. The seasons were defined as: winter (January, February, March), spring (April, May, June), summer (July, August, September), and autumn (October, November, December).”

The interpretation of the seasonality of Fig. 3b in the result section will be more detailed and used as an example of seasonality change for other figures.

19. **RC** - In the Result section, it is good to refer to Tables and Figures, but the reader also needs actual values included in the text, otherwise he always has to go back and forth from text to Table/Figure.

AC - The values were already in the text accompanying the trend interpretation, except for loads

Author's changes in manuscript:

Actual values will be added for those that were missing.

20. **RC** - 6;12: please, be more specific, and always use similar ways of describing the data: first, trends. Second, seasonal variations. It helps increasing the clarity of the manuscript and makes things easier for the reader.

AC - We described results as follows: 1) trends accompanied by actual data; 2) seasonality; 3) correlation.

Author's changes in manuscript:

This plan will be more systematically applied in the revised version

21. **RC** - You have too many additional figures. Please, make a selection of the ones that are really useful to support your ideas.

AC - Our manuscript present results of two rivers and a coastal ecosystem. We decided to place the results of one of the two rivers in supplementary materials because they must be accessible for the reader.

22. **RC** - 7;19: add a section for this correlation analysis

Author's changes in manuscript:

We will modify the description of results, rather than adding a new section (please see point #20).

23. **RC** - 7;24-29: Do you believe your DLM analysis is suitable for phytoplankton biomass description at the seasonal scale? You need to validate this first, and plots in Figure 7 don't help answer this question if you don't show residuals (you'll see that they don't look like white noise).

AC - We presume that these comments deal with phytoplankton abundances, not phytoplankton biomass. We do not understand the second sentence of the referee's comment. It is a time series and for different reasons (e.g.., missing data, irregular sampling frequencies, exceptional abundances), we believe that DLM, which has been used previously in published studies (see Soudant et al., 1997; Scheuerell et al., 2002; Hernández-Fariñas et al., 2014; Hernández-Fariñas et al., 2017) is an appropriate tool for such data. Let us define white noise as values that are mutually uncorrelated with zero mean and have the same Gaussian probability distribution.

The residuals of diatom abundances are available in Appendix 1. The residuals QQ-plot of diatom abundances is presented below.

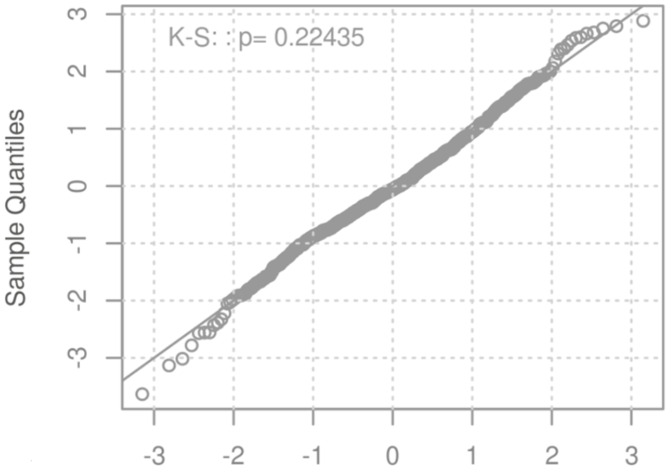

Figure R3. The QQ-plot of diatoms abundances:

The Kolmogorov-Smirnov *p*-value is equal to 0.2244. The Stoffer-Toloi (i.e., autocorrelation test) p-value is 0.1723. These results suggest that this is actually white noise.

24. **RC** - Insets in Figure 7 are not explained. It has to be.

AC - The explanation is already in the legend.

"Figure 7: Long-term trends and seasonality of Chl a (a, b), diatom (c, d), dinoflagellate (e, f) and diatom:dinoflagellate ratios (g, h) in VB. Insets show trends with optimal scale. See Fig. 2 for details"

Author's changes in manuscript:

More information on graphical representation will be added in the method section of the revised version.

25. **RC** - Section 4: this section could be reorganized as follows 1) Nutrients and Chl-a trends at river basin outlets 2) Nutrients and Chl-a trends in the bay 3) River to bay continuum 4) Implications for management

AC - The discussion section of the previous manuscript takes into account an unequivocal link between the bay and rivers, as shown in previous studies (e.g., Guillaud et al., 2008; Ménesguen and Dussauze, 2015; Ménesguen et al., 2018b). Therefore, the paragraphs described the variables (nutrients and phytoplankton) by grouping results from rivers with those from the Vilaine Bay. We understand that it was too hasty (i.e., continuum).

Author's changes in manuscript:

We propose to reorganize the discussion section as follows: 1) Eutrophication trajectories at river basin outlet 2) Eutrophication trajectories in the Vilaine Bay 3) River to Bay continuum 4) Implication for nutrient management. Please see letter to editor for detail.

26. **RC** - How does the estuarine zone could interfere in your interpretations? Same question with the presence of a dam at the outlet of the Vilaine river? This needs to be addressed, at list by listing the; different processes that occur. Many has been done on the subject.

Author's changes in manuscript:

We will add text explaining the potential influence of processes within estuaries and dam to section 4.3 of discussion (River to Bay continuum) of the revised manuscript (please see letter to editor). However, we conclude that they may not have modified trends in riverine loads.

27. **RC** - 8;21: This can't be said like this. At the outlet of large and intensively managed catchments, nutrients variations are co-controlled by upstream hydrological variations, delivery to stream modalities (point or diffuse sources?), and by instream retention processes through physical and biogeochemical processes.

AC - The lines that the referee has pointed out dealt with the variation in nutrient transfer from watershed to coastal waters, not the variations in nutrient concentrations in river waters: “The transfer of nutrients from continents to coastal waters is largely driven by freshwater inputs, the dynamics of which depend largely on precipitation in watersheds”. Thus, in our opinion, our sentence was correct.

28. **RC** - 8;22: You should mention the North Atlantic Oscillation to explain the 7 years cycles. See also Dupas et al., 2018 (WRR)

AC - The inter-annual variability (i.e., oscillation of 6-7 years) of river discharges are related to precipitation regimes, which are modulated by climate (i.e., NAO for the North Atlantic region). The relationship between flow regimes, precipitation and NAO was explicitly detailed in Radach and Pätsch (2007). We supposed that we did not need to mention this point, which is not essential to support the main subject of the paper. However, for BGS we can include reference to NAO.

Author's changes in manuscript:

The sentences corresponding to this subject will be modified as follows:

“Trends in the Loire and the Vilaine discharges displayed similar oscillations to those of rivers flowing to the North Sea as reported by Radach and Pätsch (2007), suggesting a common hydro-climatic pattern in Western Europe linked to NAO.”

29. **RC** - Figure 8 could be a great final figure, but needs to be explained once the processes explaining the different patterns in eutrophication metrics are completely described.

    Author's changes in manuscript:

    We will explain in more detail the construction of figure 8 in the text and the caption.

Technical comments

30. **RC** - Page 1, Line 14 (1;14): remove "(i.e., phytoplankton biomass)", as eutrophication expression is not only phytoplankton excessive biomass.

    Author's changes in manuscript:

    The sentence will be modified as follows:

    "The evolution of eutrophication variables (i.e., nutrients and phytoplankton biomass) during recent decades was examined in the coastal waters of the Vilaine Bay (VB, France) in relation to those in their main external nutrient sources, the Loire and Vilaine Rivers."

31. **RC** - 2;5-7: this has to do with different source types and it should be explained. Environmental measures to tackle P were successful because P largely originated from point sources with limited legacy effects in the streams. For N, diffuse sources dominate and there is large legacy effect.

    AC - The information suggested by the referee has been extensively explained in the cited articles and we found it unnecessary to add information that was detailed elsewhere. We mentioned the question of different source types and legacy effects in the discussion section 4.3.

32. **RC** - 2;7-11: you should also mention freshwater ponds and lakes were eutrophication is still severe despite large P reductions.

    AC - We can mention it, but this is not the main subject of the study. We would like to focus on eutrophication in coastal ecosystem, by highlighting the needs to reduce both P and N loads to mitigate eutrophication along the land-sea continuum.

33. **RC** - 2;16-17: I'd remove the codes for what you called "water masses". Do the authors mean "water body"?

    Author's changes in manuscript:

    The codes will be removed and the sentences will be modified (see letter to editor)

34. **RC** - 2;18: an actual scientific reference would be better.

    Author's changes in manuscript:

    The sentences will be modified and scientific references will be added in the revised version.

35. **RC** - 3;3: "widest" is not correct. You may refer to "largest river basin".

    AC - The use of the word "widest" refers to the river Loire and not to the river basin. This usage has been confirmed by our English native.

36. **RC** - 3;4: sentence is not clear, please, rephrase it.

Author's changes in manuscript:

The sentence will be rephrased as follows:

"Their catchment areas are dominated by agricultural activity, together sustaining two-thirds of the national livestock and half the cereal production."

37. **RC** - 3;22: sentence is not clear, please, rephrase it.

Author's changes in manuscript:

The sentence will be rephrased in the next version as follows.

"Nutrient and Chl a concentrations, plus phytoplankton counts in VB (Ouest Loscolo, Fig. 1) were provided by the French National Observation Network for Phytoplankton and Hydrology in coastal waters (REPHY, 2017)."

38. **RC** - 5;1: the use of ":" separates the sentence in a way that makes it hard to understand. Please, modify.

AC - Corrected

39. **RC** - 5;8-9: check for use of different tenses throughout the manuscript.

Author's changes in manuscript:

The sentence will be removed. The method section 2.4 will be detailed

40. **RC** - 5;23: Change this section title to "Discharge and nutrients long term trends in freshwater basin outlets"

Author's changes in manuscript:

The title suggested does not take into account the Chl *a*. We propose a modification to the section title as follows:

"Long term trends in eutrophication parameters in river basin outlets"

Referee 2 – Anonymous referee

Referee's comments (RC) - This paper studies the long term trends in nutrient and phytoplankton dynamics in the Loire and Vilaine rivers, and in the Vilaine Bay (VB). The authors discuss changes in eutrophication of these systems, and relate changes in the VB to those in nutrient inputs from the two rivers. They show that, even though phytoplankton blooms decreased in the riverine systems following reduction in dissolved inorganic P, phytoplankton biomass in the VB has continued to increase. This could be fueled by nitrogen delivery from the rivers (slightly increasing trend for the Loire), together with phosphorus and silica recycling from bottom sediments in the coastal area. This is an interesting discussion point, that totally fits Biogeosciences' scope. This is however only superficially discussed, and the layout of the paper makes it difficult to identify the main conclusions. I also noted important gaps in the methods' description.

The presentation of the river trajectories is extensive, but was already thoroughly discussed in a previous study (Minaudo et al., 2015). Very complete time series of Chla concentrations and abundances of different phytoplankton species in the VB are presented, and could be extremely valuable to examine changes in community structure. However, these are not discussed in depth. Moreover, more elements should be provided to the reader to justify that the data presented here is enough to support the conclusions of the study. In fact, interpretations of the dynamics in the VB are derived from observations at a single point, at which the influence of the Loire river is not obvious and not discussed.

I believe these major shortcomings should be addressed before this work can be published.

General comments

1. More information on the influence of the Loire river on the VB dynamics is needed. In fact, nutrients need to travel more than 120km from the Loire river monitoring station (Saint Luce sur Loire) to the Bay, through the Loire estuary and along the coast. Do coastal currents carry most of the Loire river's exports to the VB? How can processing in the estuary and along the coast impact loads reaching the VB?
2. Methods on the Dynamic Linear Models (DLM) and Mann-Kendall (MK) test analysis are not detailed enough. I am also not convinced that the MK test provides any more information than the DLM analysis. To my understanding, numerical estimates on trends and seasonal variations can also be extracted from the latter. Using these two methods to come up with the same interpretations waters down important messages in the results and discussion sections.
3. Authors refer several times throughout the manuscript to "management scenarios focused solely on P reduction" or on "P alone". However, this is not totally accurate for the study area, and should be moderated. Even though ecosystems responded quicker to P reduction strategies (e.g. for point sources) than to policies on agricultural fertilization, those already exist (e.g. EU Nitrates Directive).
4. In general, statements are sometimes vague or not totally accurate. The structure of the results and discussion sections makes it difficult for the reader to identify the main conclusions of the study.

These points, together with more minor concerns, are more detailed hereafter, in the specific comments.

Author's comments (AC) - We thank the referee for the detailed analysis and constructive comments. For this referee, the link between the Loire River and the VB was not well enough established in spite of numerous works cited in the previous version. We will add the information on the continuum to the revised version. In the case of the Loire/Vilaine Rivers – the VB continuum, we recognize that there was no formal decision to reduce P-only. However, the small decrease of DIN concentrations in the Vilaine and their increase in the Loire especially in summer during recent decades provide a scenario that allows testing the P-only paradigm.

We also agree to add more detail on the method used and to restructure and extend the discussion.

Concerning the influence of estuarine processes, also mentioned by referee #1, we will add texts explaining the biogeochemical processes within estuaries and dam (please see letter to editor).

All issues raised by the referee are carefully answered point by point below.

Specific comments/scientific questions

1. **RC** - L7-8, P2. "This result is consistent with the idea that reducing P alone, and not N, can mitigate eutrophication of freshwater systems (Schindler et al., 2008)": This paper from Schindler et al. does not show this; they study the effect of reducing N only. Moreover, this is not a scientific consensus (e.g. Pearl et al., 2016, Environ. Sci. Technol. 50, pp 10805–10813). This sentence should be moderated.

   AC - It is true that these authors also studied the reduction of N in their lake, but it was to support the hypothesis that the reduction of P can be enough for lake restoration. At the end of their summary, the authors also stated that to reduce eutrophication, the focus of management must be on decreasing inputs of P. In a more recent article, Schindler et al. (2016) clearly argued for a reduction of P alone to control eutrophication in lakes and other freshwater ecosystems, even though they recognize that anthropogenic nitrogen emissions can also affect human health and ecosystems (i.e., Box 2).

   Author's changes in manuscript:

   We plan to review the introduction by integrating recent articles by Schindler et al. (2016), Paerl et al. (2016) and others (see letter to editor).

2. **RC** - L14-15, P2. "Nutrient inputs ...control phytoplankton production in coastal waters of the northern Bay of Biscay": Riverine inputs constitute the major nutrient source, but don't necessarily control phytoplankton dynamics. Guillaud et al. (2018) show that sediments have a high influence on Chla levels as well (light limitation in high flow periods/winter).

   AC - It is true that there are also environmental conditions allowing nutrients to be consumed by primary producers, such as water residence time, light availability, etc. The primary production in coastal waters off the Loire and Vilaine River is limited by light availability due to insufficient irradiance during winter and suspended sediment flux from rivers and resuspension during the period of high hydrodynamic activity (Guillaud et al., 2008). However, these authors also showed that, except during periods of light limitation (November - February), phytoplankton blooms in this area respond to the variation in river discharge.

Author's changes in manuscript:

A section explaining the link between the Loire and Vilaine Rivers and the VB coastal waters will be added to the method section (please see below).

3. **RC** - L22-24, P2. Consider adding references to support this.

   Author's changes in manuscript:

   The following reference will be added (Lunven et al., 2005; Loyer et al., 2006).

4. **RC** - L9, P3. “The VB...is located under direct influence of these two rivers”: This is not really clear from Fig. 1. See general comment 1.

   AC - The link between the Loire inputs and dynamics of the coastal waters of the Northern Bay of Biscay, including the Vilaine Bay, has been established using ecological model ECO-MARS3D (see Ménesguen et al., 2018b). These authors showed in their figure 6 the influence area of several large French Atlantic river plumes during three different flow regimes. It also showed that the VB coastal waters are always affected by the Loire river plume whatever the regime scenario. This can justify the link between the Loire River and the VB (i.e., continuum).

   Author's changes in manuscript:

   The following reference will be added to support the sentence mentioned by referee (Guillaud et al., 2008; Ménesguen and Dussauze, 2015; Ménesguen et al., 2018b).

   The section 2.1 will be completed with the following text explaining the contribution of the Loire and Vilaine Rivers to the VB fertilization.

   “The Loire river plume tends to spread north-westward with a dilution of 20 to 100-fold by the time it reaches the VB (Ménesguen and Dussauze, 2015; Ménesguen et al., 2018b). The Vilaine river plume tends to spread throughout the bay and then move westward (Chapelle et al., 1994). The ECO-MARS3D model estimates that the Loire constitutes >60% of VB DIN concentrations during flood regimes and from 20 to 40% during low discharge periods (Gohin, 2012; M. Plus, comm. pers.). Exceptional floods from the Loire and Vilaine can lead to high surface water turbidity in the VB (Guillaud et al., 2008).”

5. **RC** - L4-5, P4. The link between the first two sentences of this paragraph is not clear.

   Author's changes in manuscript:

   These sentences will be modified as follows.

   “DIN was defined as the sum of nitrate, nitrite and ammonium, with nitrate as major component (>90%).”

6. **RC** - L25, P4 – L6, P5. This paragraph would benefit from more explanations on the DLM method. When you say “look like interpolation”, do you mean it is equivalent to interpolation? If yes, which kind of interpolation?

   AC - The sequential DLM approach is provided by the Kalman filter, by identifying the missing values and replacing them with normal random variables. This approach may be viewed as one that uses a prior for the parameter which replaces the missing values. This is another way to say “absence of data leads to no change in distributions for model parameters”.

Author's changes in manuscript:

This method section 2.4 will be reorganized and the sentence corresponding to this subject will be replaced with the following:

The DLM approach is particularly suitable for environmental data series characterized by outliers, irregular sampling frequency and missing data. The latter are taken into account by the Kalman filter (Kalman, 1960), a component of the DLM, i.e., no information leads to no changes in distributions for model parameters (West and Harrison, 1997). This approach may be viewed as one which uses a prior for the parameter which replaces the missing value.

RC - Why do you choose to fit second order polynomial functions for the trends, and bimodal trigonometric functions for the seasonality? Is it based on any preliminary analysis of the data?

AC - We choose a second order polynomial model because looking at the log-transformed time series it appeared to us that a first order (i.e., adapting trend up to linear) was too restrictive and a third order (i.e., adapting trend up to cubic) was not necessary, leading to an over fitted model.

In our geographical area, the annual patterns of phytoplankton variability have a six months periodicity as described by Winder and Cloern (2010). This bimodal pattern is characterized by two peaks per year, such as spring and autumn or summer and winter blooms. In order to allow our model to adapt to such periodicity we have to include a two harmonics seasonal component. Thus, yes it is based on preliminary analysis of the data.

Author's changes in manuscript:

A justification of this subject will be added in the section 2.4 of revised manuscript.

RC - What does “time units” refer to? Is it the frequency at which the trends/seasonal variations are estimated?

AC - Time unit is the smallest time interval between sampling dates within a period of analysis. In our case, the period is one year. Time units are weekly, fortnightly or monthly depending on the data. Seasonal variations are estimated for each time unit.

RC - Why are those plotted with (two different types of) log scales? It makes it more difficult to link the figures with the values provided in text.

AC - Y-axis of all graphics has been modified to show original units using a log-scaled y-axis.

7. **RC** - L14-19, P5. What extra information does the MK test provide? Trend values can already be extracted from the DLM analysis. Is the method applied to the trend/seasonality functions from the DLM analysis, or to the raw data? Are uncertainties accounted for?

AC - The modified MK test was used as a formal trend significance test. The test was applied respectively to trend and seasonality components from DLM, not to the raw data (please see comment from referee #1point #7 for further detail).

Yes, uncertainties are taken into account by the DLM.

Author's changes in manuscript:

The information on the modified Mann-Kendall test will be more detailed and clearly explained in the revised version.

8. **RC** - L19, P7-L7, P8. Results on Chla concentrations and phytoplankton species in the VB are not thoroughly presented here. It seems from the seasonality plot that, in the timeframe of the study, Chla has always peaked in spring and summer, and that since 2006 the summer peak has reached similar concentrations to the spring one. It's also interesting to note that there seems to be a succession of 3 algae blooms: a diatom bloom in spring, a dinoflagellate one in early summer, when DSi is depleted, and another diatom one in late summer.

   AC - In this study, we used only the total counts of diatoms and dinoflagellates, to account for the role DSi. These two groups represent >85% of total micro-phytoplankton counts and thus the biomass (section 2.3, line 9-12, page 4).

   This change in Chl *a* seasonality was mainly due to the increase in summer diatom abundances and the decrease in spring ones, as suggested by their seasonality.

   The seasonal pattern of phytoplankton blooms was characterized by a diatom bloom in spring corresponding to high river flows and another one in late summer. Dinoflagellates tend to increase in summer but their abundances remain lower than those of diatoms, except during discolored water events (Souchu et al., 2013; Sourisseau et al., 2016). The collapse of spring diatom bloom is due more to DIP depletion than DSi (please see point #11).

9. **RC** - L21-22. P8. Why would trends in discharge in the studied rivers depend on variations in the precipitation in river basins flowing to the North Sea?

   Author's changes in manuscript:

   It will be rephrased as follows:

   "Trends in the Loire and the Vilaine discharges displayed similar oscillations to those of rivers flowing to the North Sea as reported by Radach and Pätsch (2007), suggesting a common hydro-climatic pattern in Western Europe linked to NAO."

10. **RC** - L30. P8-L4. P9. This paragraph would be more convincing if estimates of the loads from the different sources were provided. Is the Loire "probably" the major nutrient source, or has it been shown that it actually is? How much water/nutrients are retained in the Arzal dam, and how does it influence the loads reaching the VB? Are the discharge and loads from the Vilaine really negligible in summer, even though it flows directly to the Bay, while the Loire river plume has to travel 120km?

    AC - According to the modelling study, the Loire is actually the major nutrient source (please see point #4).

    Concerning the Vilaine, during the period of low water discharge (10 – 100 m^3 s^{-1}), the dam is closed at high tide. The small releases due to the lock functions of the dam (shipping, fish-way and salt-water pump) represent half of the "natural" discharge (Traini et al., 2015). The dam is closed below 10 m^3 s^{-1}. The summer Vilaine discharge measured at Rieux displayed strong variations, ranging between <1 to 100 m^3 s^{-1}, with >95% of values below 60 m^3 s^{-1}. Therefore, we consider that half of water discharge during the low water period was retained by the dam.

Unfortunately, we do not have any measurement of nutrient concentrations at the dam outlet nor inside the dam, which could be used to estimate the nutrient retention. The presence of a dam at the river outlet may increase water residence time, thus favor nutrient uptake by phytoplankton and loss of N via denitrification (Howarth et al., 1996; Seitzinger et al., 2006). The presence of Arzal dam may thus attenuate nutrient transport from the Vilaine River to the VB. The use of loads calculated from Rieux station has likely overestimated nutrient loads from the Vilaine River. However, taking into account dam retention for the Vilaine, our estimate of nutrient loads from the Loire remains higher than those from the Vilaine (see Table 4).

Author's changes in manuscript:

The estimates of loads from the Loire and Vilaine Rivers will be added to the text.

Concerning the influence of estuarine processes, we provided text that will be added to the revised manuscript (please see letter to editor).

11. **RC** - L9. P9-L20. P9. The phytoplankton succession is not thoroughly discussed here. See Specific comment 8. Even though they are decreasing, spring diatom abundances are still superior to summer ones. It is mentioned that temperature changes can induce shifts in species' succession. Is it the case here? It would also be interesting to discuss the relationship between phytoplankton successions and variations in DSi, for example.

    AC - Concerning phytoplankton succession, please see point #8. This is true that the spring diatom abundances remain higher than summer ones. The point that we would like to highlight here is the increase in summer diatom abundances, accompanying the increase in summer Chl a as the indicator of the VB degradation. The increase in sea surface temperature (SST) has been reported in the Bay of Biscay by Huret et al. (2013). Désaunay et al. (2007) have reported an increase in winter temperatures on the continental shelf. Thus, the change in the timing of annual maxima observed for phytoplankton biomass could not be attributed to the increase in temperature and was better explained by changes in nutrient loads. Moreover, changes in phytoplankton community structure at species level, in relation to changes in environmental parameters (e.g., temperature), will be examined in other study.

    In the coastal waters off the Loire and Vilaine Rivers, including the VB, the phytoplankton bloom is generally limited by DIP in spring and by DIN in summer (Loyer et al., 2006; Guillaud et al., 2008). This pattern has been verified by bioassay (M. Retho, Ifremer 2015, unpublished data). On the basis of DIP and DSi concentrations, and DSi:DIP ratios, the diatoms are rarely limited by DSi. The decrease in DIP riverine loads has increased the DSi:DIP ratios in the VB during the past decades (Fig. S7) and reinforced therefore the DIP limitation in the VB, as suggested by Billen et al. (2007) for the Seine River – the Seine Bay continuum.

    Author's changes in manuscript:

    The discussion section will be reorganized (please see letter to editor). We will discuss in more detail the relationship between diatom/dinoflagellate dynamics and variations in DSi concentrations.

12. **RC** - L20. P10. Does Table S2 show values for the Bay of Biscay or for the Ouest Loscolo station only?

AC - No, it shows global annual median values for the Bay of Biscay, as detailed in the table legend.

13. **RC** - L21-23. P10. Precise that these correlations are at the annual scale. Seasonal variations of DIN and DSi do not seem correlated.

    AC - The precision of correlation analysis has been given in the end of the section 2.4. We explained that we used annual median values to compute Spearman's Rank correlation analysis.

    As mentioned in Table 3, annual medians of DIN and DSi in the VB were correlated.

    Author's changes in manuscript:

    The sentence will be modified as follows.

    "Significant negative correlations between annual medians of Chl a in the VB and Chl a and DIP in rivers, as well as significant positive correlations between annual medians of DIN and DSi in the VB with those of river discharge suggest that changes in eutrophication parameters in the VB (i.e., phytoplankton biomass) were directly related to changes in rivers."

14. **RC** - L5-16. P11. Please provide some numbers to support your conclusions.

    Author's changes in manuscript:

    Numbers will be added.

15. **RC** - L9-12. P12. An opening on eutrophication and its mitigation would fit better. regarding the introduction.

    AC - The conclusion section will be modified following modification made in discussion section.

    Author's changes in manuscript:

    The text below will be added concerning perspective on eutrophication mitigation

    The internal loads of nutrients from sediments may counteract the reduction of external nutrient loads and may delay the restauration progress. Taking into account these internal processes in modelling studies (i.e., ECO-MARS3D model, Ménesguen et al., 2018a, b; Menesguen and Lacroix, 2018) will better simulate nutrient load reduction scenarios.

16. Table S1. When different measurement methods were used for a same variable. consider indicating which time period corresponds to which method.

    Author's changes in manuscript:

    This information will be added to the revised manuscript, particularly for dataset in the VB.

Wording

RC - Throughout the text: "Vilaine Bay/VB" -> "the Vilaine Bay/VB"

AC - Corrected

RC - L15. P1. "in relation to those in their..." -> "in relation to changes in its"?

AC - Corrected

RC - L4. P2. “myriad responses” -> “myriad of responses”

AC - The use of “myriad responses” was based on Cloern (2001) and it has been validated by our English native.

RC - L15-18. P4. “The removed...general trend observed”: Please reformulate.

AC - Corrected

RC - L10&12. P5. “position of” -> “timing of”

AC - Corrected

RC - L14. P9 & L8. P12. “course” -> “succession

AC - Corrected

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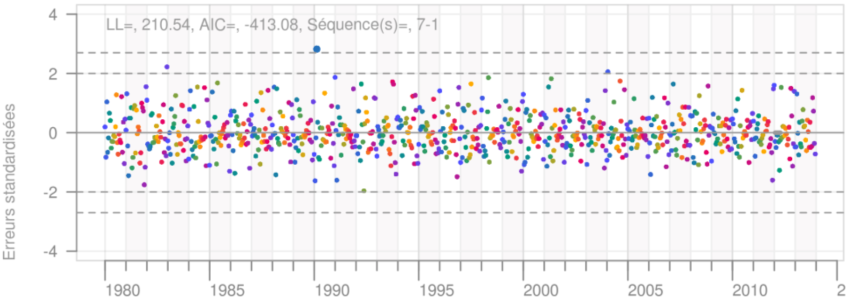
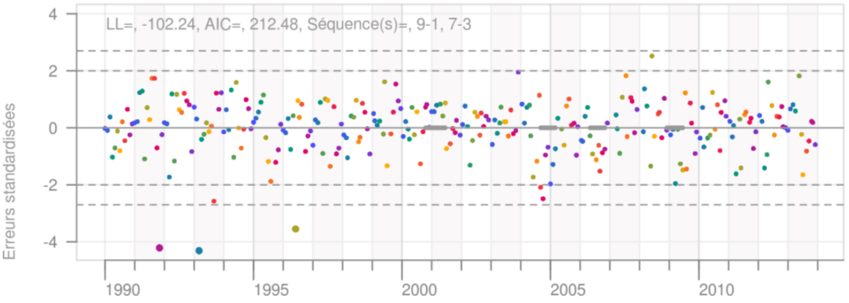
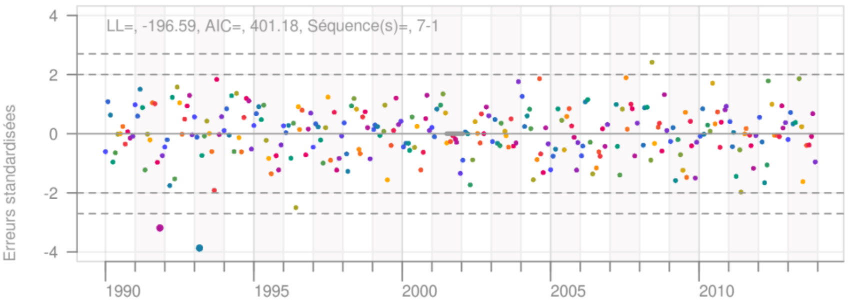
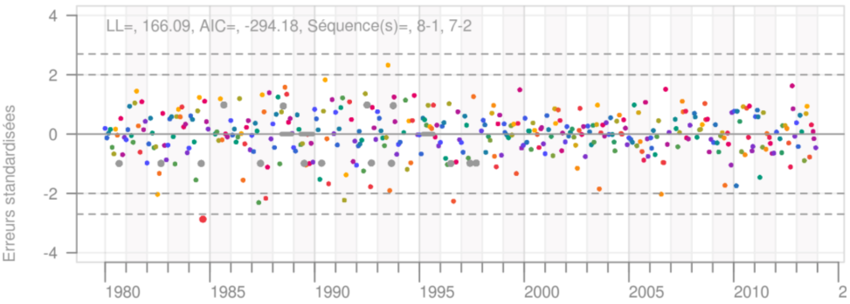
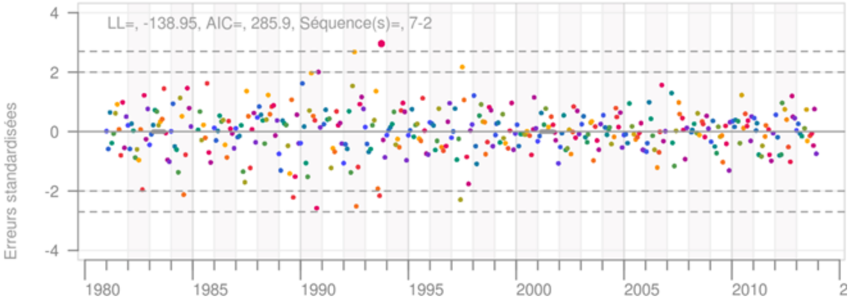
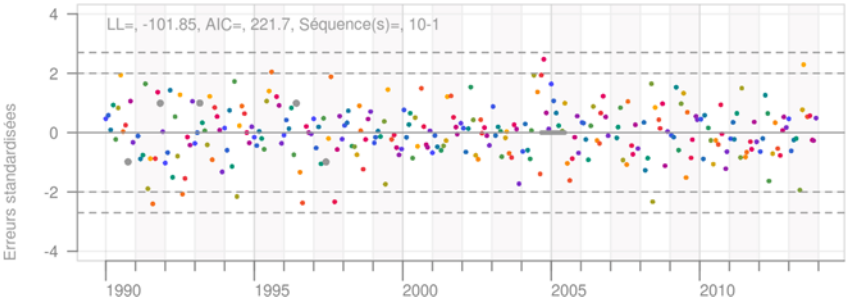
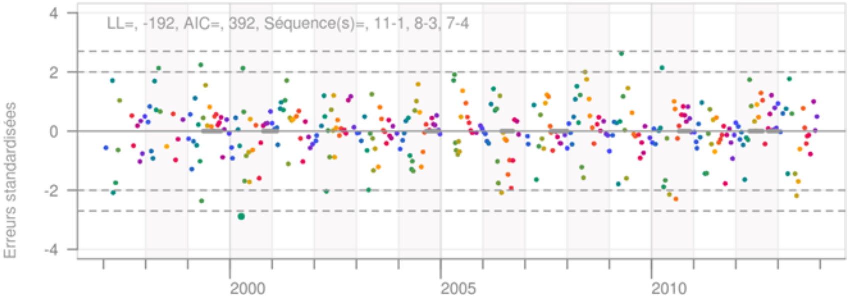
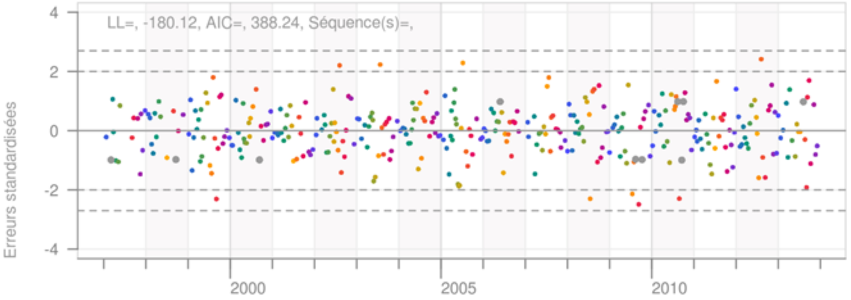
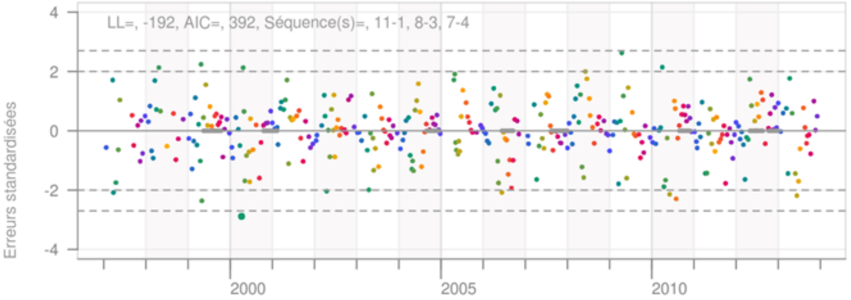
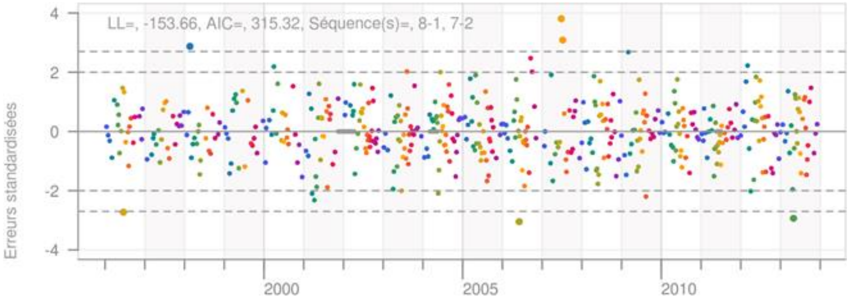
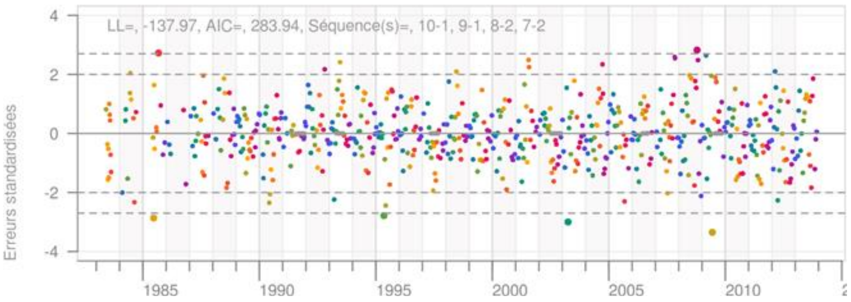
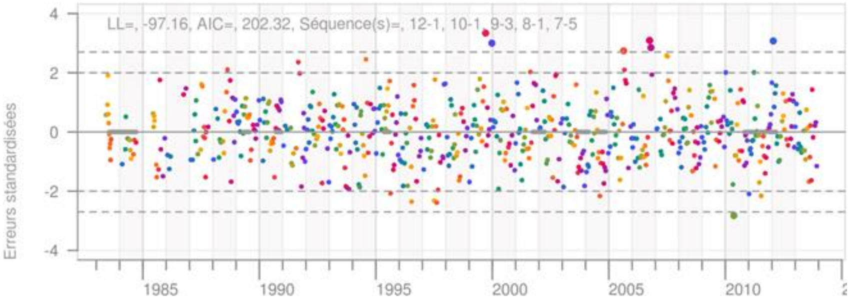
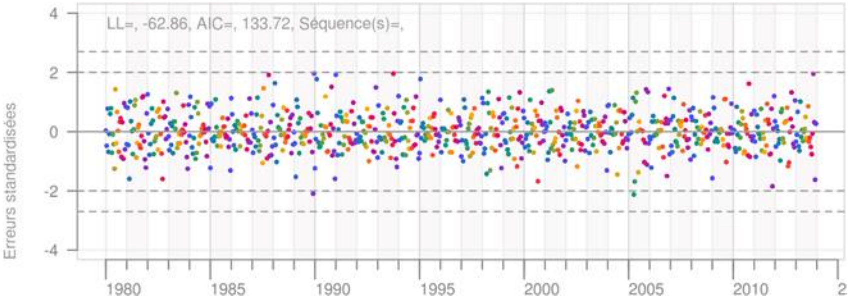
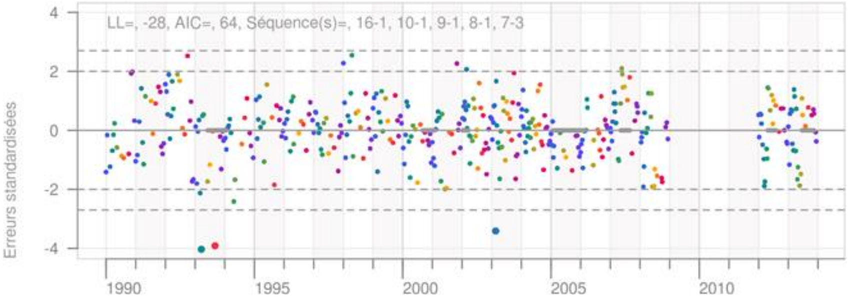
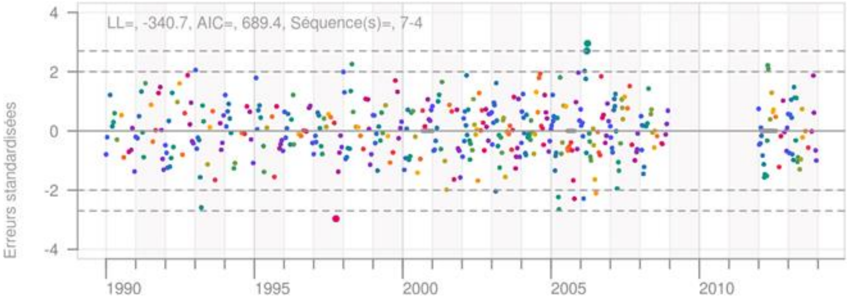
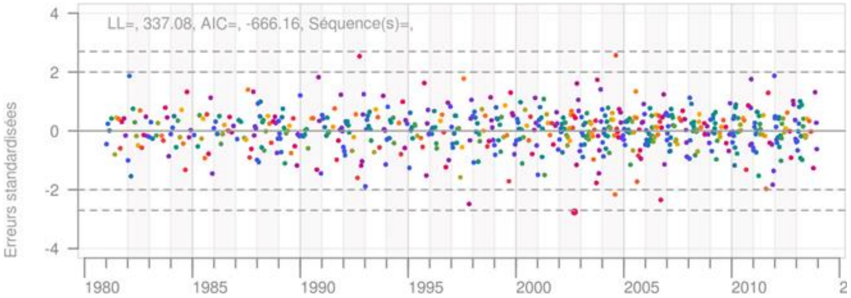
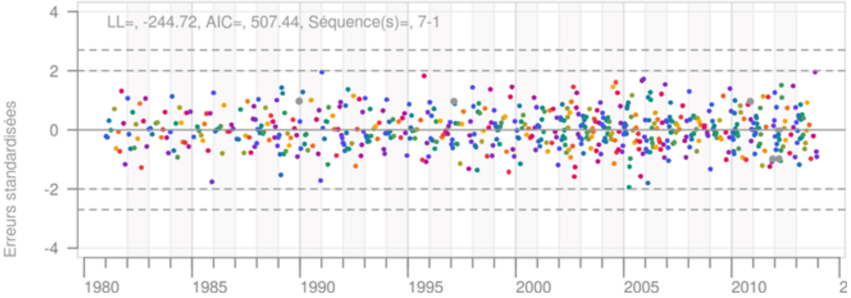
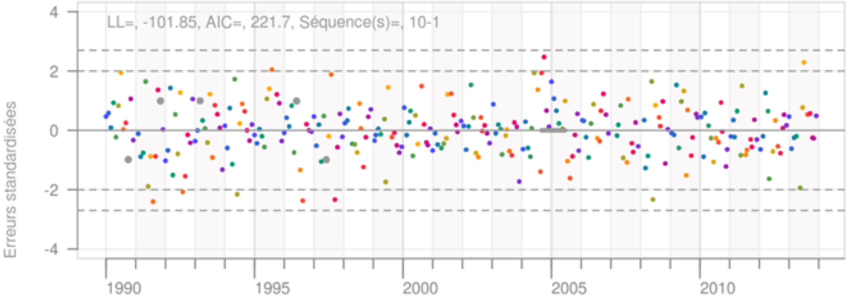
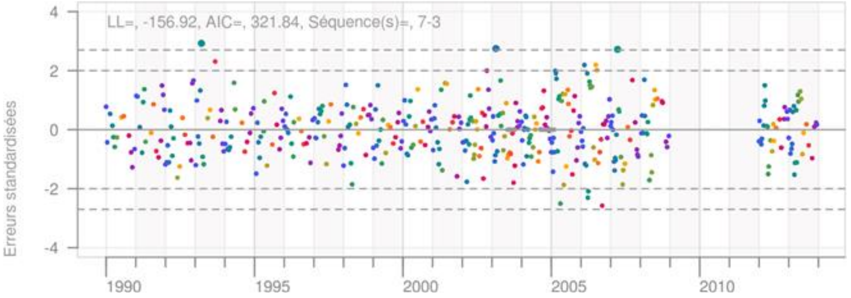
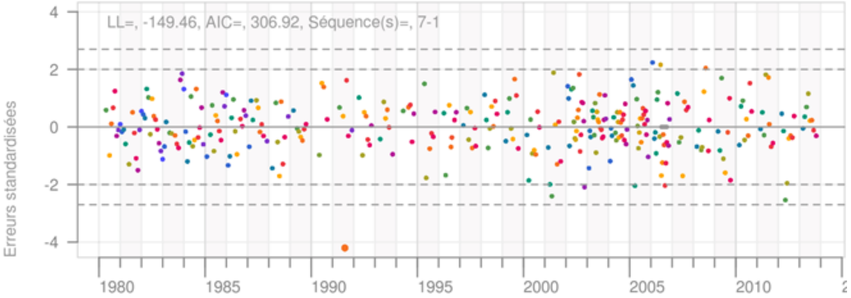
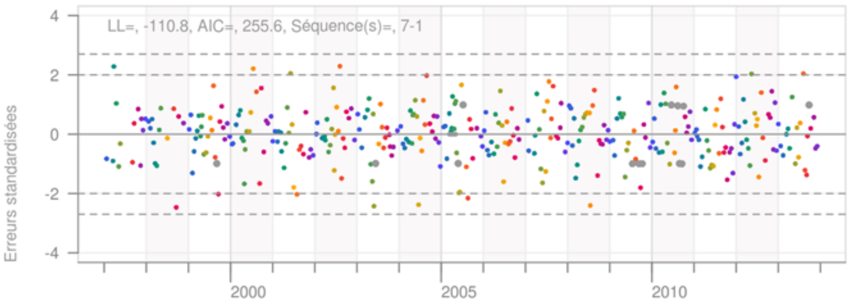
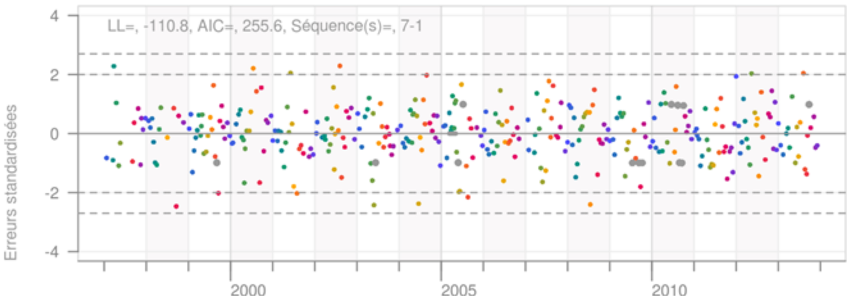
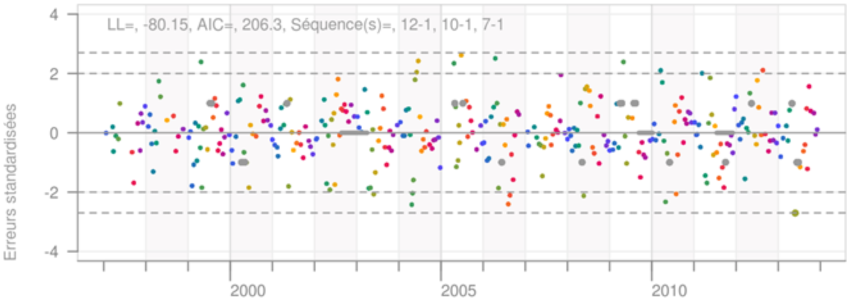
Appendix 1. Residual plots for all parameters used in the present study

Figures	**Parameters**	**Residual plots**
2	Loire discharge	
3	Loire DIP	
3	Loire DIP Loads	
4	Loire DIN	
4	Loire DIN Loads	

Figures	Parameters	Residual plots
5	Loire Chl *a*	
6	Vilaine Bay DIP	
6	Vilaine Bay DIN	
6	Vilaine Bay DSi	
7	Vilaine Bay Chl *a*	

Figures	Parameters	Residual plots
7	Vilaine Bay Diatom	
7	Vilaine Bay Dinoflagellate	
Supp 2	Vilaine discharge	
Supp 3	Vilaine DIP	
Supp 3	Vilaine DIP loads	

Figures	Parameters	Residual plots
Supp 4	Vilaine DIN	
Supp 4	Vilaine DIN loads	
Supp 5	Loire DIN/DIP	
Supp 5	Vilaine DIN/DIP	
Supp 6	Vilaine Chl a	

Figures	Parameters	Residual plots
Supp 7	Vilaine Bay DIN/DIP	
Supp 7	Vilaine Bay DIN/DSi	
Supp 7	Vilaine Bay DSi/DIP	

Reduced phosphorus loads from the Loire and Vilaine Rivers were accompanied by increasing eutrophication in Vilaine Bay (South Brittany, France)

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Abstract. The evolution of eutrophication parameters (i.e., nutrients and phytoplankton biomass) during recent decades was examined in ~~the~~ coastal waters of the Vilaine Bay (VB, France) in relation to those in ~~their main external nutrient sources,~~ the Loire and Vilaine Rivers. Dynamic Linear Models~~, corroborated by the Mann-Kendall test,~~ were used to study long-term trends and seasonality of dissolved inorganic nutrient and chlorophyll *a* concentrations (Chl *a*) in rivers and coastal waters ~~between 1980 and 2013~~. For the period 1997-2013, ~~The~~ the reduction in dissolved riverine inorganic phosphorus concentrations (DIP) ~~from the early 1990s~~ led to the decrease in their Chl *a* levels. However, while dissolved inorganic nitrogen concentrations (DIN) decreased only slightly in the Vilaine, ~~and actually~~they increased in the Loire, ~~especially during~~specifically in summer. Simultaneously, phytoplankton in the VB ~~has~~ under~~gone~~went profound changes with~~: 1)~~

increase in biomass~~, 2)~~ and change in the ~~position~~ timing of the annual peak from spring to summer~~, and 3) increase in diatom:dinoflagellate ratios, especially in summer~~. The increase in phytoplankton biomass in the VB, manifested particularly by increased ~~in~~ summer diatom abundances, was ~~probably~~ due to ~~increased~~ enhanced summer DIN loads from the Loire, sustained by internal regeneration of DIP and dissolved silicate (DSi) from sediments. ~~This~~ The long-term trajectories of this case study provide a more evidence that significant reduction of P inputs without simultaneous N abatement was not yet sufficient to control eutrophication all along the Loire/Vilaine – VB continuum~~ecosystem-scale analysis reports the consequence of nutrient management scenarios focused solely on P reduction~~. ~~Freshwater ecosystems u~~Upstream rivers reveal indices of~~successful~~ recoveries ~~through~~ following the ~~control~~ significant diminution of P~~ alone~~, while eutrophication continues to increase downstream, especially during the period of N limitation. ~~Therefore, nutrient management strategies~~More N input reduction, paying particular attention to diffuse N-sources, ~~are~~ is required to control eutrophication in receiving VB coastal waters. Internal benthic DIP and DSi recycling appears to have contributed to the worsening of summer VB water quality, augmenting the effects of anthropogenic DIN inputs. For this coastal ecosystem, nutrient management strategies should consider the internal nutrient loads in counteracting decreased external inputs.

Keywords: eutrophication, ~~nutrients,~~ phytoplankton, internal nutrient loads, dual nutrient reductions, Vilaine Bay, Dynamic Linear Models

1 Introduction

Anthropogenic eutrophication is widely regarded as one of the major problems affecting both inland and coastal aquatic ecosystems (Downing, 2014). The increase in phytoplankton biomass is the most common symptom of eutrophication among the myriad responses of aquatic ecosystems to anthropogenic inputs of nitrogen (N) and phosphorus (P) (Cloern, 2001; Glibert et al., 2011). Since the beginning of the 1990s, measures to reduce nutrient inputs in European rivers were more effective for P, originating largely from point sources, than for N, coming mainly from diffuse sources (Grizzetti et al., 2012). However, this strong imbalance between N and P input reduction ~~but~~ still led to ~~reduction~~ substantial decrease in phytoplankton biomass in many European rivers (Istvánovics and Honti, 2012; Romero et al., 2013). This result is consistent with the idea that P universally limits primary productivity in many freshwater ecosystems (Correll, 1999). Thus, reducing P ~~alone~~inputs, and not N, can mitigate eutrophication of freshwater ecosystems (Schindler et al., 2008; Schindler et al., 2016).

Despite significant P input reduction, eutrophication persists in some rivers (Neal et al., 2010; Bowes et al., 2012; Jarvie et al., 2013), and particularly in downstream coastal ecosystems, where the primary productivity is often limited by N (Ryther and Dunstan, 1971; Howarth and Marino, 2006; Paerl, 2018). As freshwater systems drain into coastal waters (Vannote et al., 1980; Bouwman et al., 2013), the efficient P reduction without simultaneous N abatement may result in more N being transported downstream, where it can exacerbate eutrophication problems in coastal ecosystems, delaying recovery (Paerl et al., 2004).~~However, this strategy does not take into account downstream coastal ecosystems, some of which evidence a slow restoration progress,~~ for example the Neuse River Estuaries (Paerl et al., 2004), the Chesapeake Bay (Harding et al., 2016), Belgian coastal waters ~~,~~ (Lancelot et al., 2007), and the Seine Bay (Romero et al., 2013) ~~and many other coastal ecosystems (see Cloern, 2001)~~. Despite more than 20 years of nutrient reduction implementation in European freshwater ecosystems, including rivers (e.g., Nitrates Directive, 91/676/EEC; Urban Waste Water Treatment Directive, 91/271/EEC), little measurable progress has been observed in many European coastal waters (EEA, 2017; OSPAR, 2017).

The Loire River, alongside the Vilaine River, ~~is part of~~are among these major European rivers whose phytoplankton biomass and P concentrations have decreased since the early 1990s, but with~~out~~ minor, if any, simultaneous ~~a~~ diminution in N concentrations (Romero et al., 2013; Minaudo et al., 2015). Affected by the Loire and Vilaine river runoff (Guillaud et al., 2008; Gohin, 2012; Ménesguen et al., 2018b), ~~Nutrient inputs from these two rivers control phytoplankton production in coastal waters of the northern Bay of Biscay (Loyer et al., 2006; Guillaud et al., 2008). According to the criteria established within the European Water Framework Directive, the ecological status of coastal waters off the Loire and Vilaine Rivers, corresponding to water masses FRGC44, FRGC45 and FRGC46, are classified as damaged areas due to elevated phytoplankton biomass (http://envlit.ifremer.fr/var/envlit/storage/documents/atlas_DCE/scripts/site/carte.php?map=LB). Among these water masses,~~ the Vilaine Bay (VB~~; FRGC44~~) is one of the European Atlantic coastal ecosystems most sensitive to eutrophication (Chapelle et al., 1994; Ménesguen et al., 2014, 2019). The VB coastal waters are classified as a problem area due to elevated phytoplankton biomass, according to the criteria established within OSPAR (OSPAR, 2017) and the European Water Framework Directive (Ménesguen et al., 2018b). ~~The VB has already undergone bottom anoxia accompanied by massive mortalities of fish in summer 1982 (Rossignol-Strick, 1985).~~ However, there is little information on how eutrophication parameters have evolved in the VB over the past 20 years in the light of eutrophication mitigation in the Loire and Vilaine Rivers. An approach taking into account seasonal variations is required as phytoplankton in many coastal ecosystems, such as the coastal waters off the Loire and Vilaine Rivers ~~VB~~, is ~~usually~~ often limited by P in spring and by N in summer (Lunven et al., 2005; Loyer et al., 2006).

In temperate coastal waters, diatoms and dinoflagellates constitute the two dominant phytoplankton classes (Sournia, 1982; Sournia et al., 1991). In term of nutrient requirements, the balance between these classes is controlled by silica (Si) availability. Increased inputs in N and P (and not Si) in aquatic ecosystems can lead to limitation in diatom biomass due to lack of dissolved silicate (Conley et al., 1993). Therefore, increasing eutrophication may favor the development of non-siliceous algae, such as dinoflagellates and harmful species (Billen and Garnier, 2007; Lancelot et al., 2007; Howarth et al., 2011).

The present study ~~examined~~ investigated the long-term ~~changes~~ evolution (trend and seasonality) of eutrophication parameters (dissolved inorganic nutrient concentrations and phytoplankton biomass) ~~and nutrient concentrations in the coastal waters of~~in the VB coastal waters, in relation to those in the Loire and the Vilaine between 1980 and 2013, using Dynamic Linear Models~~ and the non-parametric Mann-Kendall test~~. This long-term ecosystem-scale analysis provided an opportunity to test the hypothesis that eutrophication trajectories in the downstream VB coastal waters during recent decades have been influenced by those in the Loire and Vilaine Rivers. We aim to establish the link between fresh and marine water trajectories and highlight the impact of nutrient reduction strategies in rivers on coastal water quality.~~the reduction of P alone in inland aquatic ecosystems is sufficient to mitigate eutrophication in coastal waters.~~

2 Material and Methods

2.1 Sites

The Loire is the longest and widest river in France (1,012 km) with a watershed of 117,000 km^2, while the Vilaine watershed is only 10th the size, with an area of 10,800 km^2 (Fig. 1). Their catchment areas are ~~predominantly~~ dominated by agricultural activity, together ~~producing~~ sustaining two-thirds of the national livestock and half the cereal production (Bouraoui and Grizzetti, 2008; Aquilina et al., 2012). The Arzal dam, 8 km from the mouth of the Vilaine, was constructed in 1970 to regulate freshwater discharge and prevent saltwater intrusion (Traini et al., 2015). The two studied rivers, especially the Loire, are the main nutrient sources in the northern Bay of Biscay, including VB (Guillaud et al., 2008; Ménesguen et al., 2018a).

The VB, average depth 10 m, is located under direct influence of these two rivers (Fig. 1). The Loire river plume tends to spread north-westward with a dilution of 20 to 100-fold by the time it reaches the VB (Ménesguen and Dussauze, 2015; Ménesguen et al., 2018b). The ECO-MARS3D model estimates that the Loire constitutes >60% of VB DIN concentrations during flood regimes and from 20 to 40% during low discharge periods (Gohin, 2012; M. Plus, Ifremer Brest, pers. comm.). The Vilaine river plume tends to spread throughout the bay before moving westward (Chapelle et al., 1994).

The water residence time in the ~~bay~~ VB varies between 10 and 20 days depending on the season and tends to be longer during calm periods (Clément, 1986; Chapelle, 1991), with tidal ranges varying between 4 and 6 m (Merceron, 1985). The water circulation is characterized by low tidal and residual currents, driven mainly by tides, winds and river flows (Lazure and Salomon, 1991; Lazure and Jegou, 1998). During periods of prevailing winds, particularly from south-west and west, the water column of the VB is subjected to vertical mixing, which can lead sometimes to sediment resuspension and high turbidity (Goubert et al., 2010). Except during winter and period of high hydrodynamic activity, phytoplankton production in the VB is not limited by light (Guillaud et al., 2008).

2.2 Long-term monitoring dataset: Rivers~~,~~ and VB~~ilaine Bay, Bay of Biscay~~

The Loire-Brittany River Basin Authority (http://osur.eau-loire-bretagne.fr/exportosur/Accueil) furnished dissolved inorganic nutrients and phytoplankton biomass data (dissolved inorganic phosphorus concentrations, DIP; dissolved inorganic nitrogen concentrations, DIN, dissolved silicate concentrations, DSi and chlorophyll *a* concentrations, Chl *a*) in rivers, at pre-estuarine stations located closest to the river mouth upstream of the haline intrusion (Fig. 1). DIN was defined as the sum of nitrate, nitrite and ammonium, with nitrate as the major component (>90%). Sainte-Luce-sur-Loire on the Loire and Rieux on the Vilaine provided DIP, DIN and Chl *a*, measured monthly since the 1980s. For Sainte-Luce-sur-Loire, the influence of tidal dynamics was avoided by discarding data collected during high tide. Monthly DSi data were available from 2002 at Montjean-sur-Loire on the Loire and at Férel on the Vilaine (Fig. 1).

In order to calculate riverine nutrient loads, gauging stations located close to the river mouth were selected. River discharge data were extracted from the French hydrologic “Banque Hydro” database (http://www.hydro.eaufrance.fr/). For the Loire, river discharge measurements at Montjean-sur-Loire were used due to the absence of data at Sainte-Luce-sur-Loire. For the Vilaine, daily discharge data were available at Rieux from the 1980s. DIN and DIP loads from rivers were calculated using averaged monthly discharge and individual monthly nutrient concentrations (Romero et al., 2013).

Nutrient and Chl *a* concentrations, plus phytoplankton count data in the VB, provided by the French National Observation Network for Phytoplankton and Hydrology in coastal waters (REPHY, 2017), were collected from Ouest Loscolo station (Fig. 1). This station is representative of the VB coastal waters (Gohin, 2011; Bizzozero et al., 2018; Ménesguen et al., 2019) and displayed the longest dataset (from 1983 for phytoplankton counts and 1997 for nutrient and Chl *a* concentrations).~~River discharge measurements at Montjean-sur-Loire on the Loire and Rieux on the Vilaine (Fig. 1) were extracted from the French hydrologic “Banque Hydro” database (http://www.hydro.eaufrance.fr/). These two gauging stations serve as reference for calculating river inputs (Romero et al., 2013; Ménesguen et al., 2018b). Nutrients and phytoplankton biomass data (dissolved inorganic phosphorus concentrations, DIP; dissolved inorganic nitrogen concentrations, DIN, dissolved silicate concentrations, DSi and chlorophyll *a* concentrations, Chl *a*) in rivers were furnished by the Loire-Brittany River Basin Authority (http://osur.eau-loire-bretagne.fr/exportosur/Accueil). DIP, DIN and Chl *a* data came from Sainte-Luce-sur-Loire on the Loire and Rieux on the Vilaine, and DSi from Montjean-sur-Loire on the Loire and Férel on the Vilaine~~

~~(Fig. 1). Nutrient and Chl *a* data, plus phytoplankton counts (see below) VB collected at Ouest Loscolo (Fig. 1), were provided by the French National Observation Network for Phytoplankton and Hydrology in coastal waters (REPHY, 2017).~~

Acquisition periods, sampling frequencies and methods of analysis are ~~presented~~ detailed in Table S1. Briefly, nutrient concentrations were measured manually or automatically in flow analysis using standard colorimetric methods with fluorimetry or photometry detection. Chlorophyll *a* concentrations (Chl *a*) were measured with either spectrophotometry or fluorimetry. Microscopic quantitative micro-phytoplankton analyses in coastal waters were conducted on Lugol-fixed samples and counted according to Utermöhl (1958). Phytoplankton identification and ~~counting~~ counts were carried out ~~essentially~~ for organisms whose size is >20 µm (i.e., micro-phytoplankton)~~, also~~ and smaller species with chain structure ~~or colony forming~~. Further details about sampling and processing of phytoplankton species are available in ~~Lefebvre et al. (2011),~~ Hernández-Fariñas et al. (2014) and Belin and Neaud-Masson (2017). In order to account for the role of DSi, of all the micro-phytoplankton classes, genera and species identified in the VB, only total counts of diatoms (Bacillariophyceae) and dinoflagellates (Dinophyceae) were used in this work. Other micro–phytoplankton classes (Dictyophyceae, Prasinophyceae, Cyanophyceae, Chrysophyceae and Raphidophyceae) together represented only 10 to 15 % of the VB total counts (Belin and Soudant, 2018).

2.3 Time-series analyses

2.3.1 Data pre-processing

Prior to analysis, all datasets were examined using time scaled scatter plots. For DIP in rivers, these showed periods during which a limited set of values appeared repeatedly (Fig. S1), which resulted from analytical problems (Loire-Brittany River Basin Authority, S. Jolly, pers. comm.). Consequently, these suspect data were discarded to avoid misinterpretation. The removed DIP datasets represented 29% and 31% of the total number of data, corresponding respectively to the period 1980-1989 in the Loire, and 1980-1989 and 2009-2011 in the Vilaine. DSi in rivers was not analyzed for trends because of the short data period.

Prior to time series decomposition, a variance-stabilizing base *e* log transformation was applied to all variables, except for phytoplankton counts for which the base was 10, to ensure compliance with the constant variance assumption (i.e. homoscedasticity). ~~Most of DIN concentrations measured in fresh and marine waters consisted mainly of nitrate (>90%; Garnier et al., 2010; Ménesguen et al., 2018b). Therefore, DIN, defined as the sum of nitrate, nitrite and ammonium, was used for trend analysis. DIN and DIP loads from rivers were calculated using averaged monthly discharge and individual monthly nutrient concentrations (Romero et al., 2013). DSi in rivers was not analyzed for trends because of the short data period (none before 2002). Dataset in the Bay of Biscay were not included for trend analysis due to the short periods and irregularity of sampling. Of all the micro–phytoplankton classes, genera and species identified in Vilaine Bay, only total counts of diatoms (Bacillariophyceae) and dinoflagellates (Dinophyceae) were used in this work to account for the role of DSi. Other micro–phytoplankton classes (Dictyophyceae, Prasinophyceae, Cyanophyceae, Chrysophyceae and Raphidophyceae) together represented only 10 to 15 % of total counts. Prior to analysis, all datasets were examined using time scaled scatter plots. Only for DIP, it showed periods during which a limited set of values appeared repeatedly (Fig. S1), which were due to analytical problem (Loire-Brittany River Basin Authority, S. Jolly, pers. comm.). Consequently, these suspect data were not taken into account to avoid misinterpretation. The removed DIP datasets represented 29% and 31% of the total number of data, corresponding to respectively the period 1980-1989 in the Loire, and 1980-1989 and 2009-2011 in the Vilaine, but not change the general trend observed.~~

2.~~4~~ 3.2 Time-series decomposition~~analyses~~

The time-series were modeled using Dynamic Linear Models (DLM, West and Harrison, 1997) with the *dlm* package (Petris, 2010) in R software (R core team 2016). This tool belongs to the family of methods which encompass, for example, State-Space models, Structural Time Series Model, Unobserved Component Model (Harvey et al., 1998) and Dynamic Harmonic Regression (Taylor et al., 2007). The model decomposes an observed time-series into component parts, typically trend, seasonal component (i.e., seasonality) and residual. The DLM approach is particularly suitable for environmental data series characterized by outliers, irregular sampling frequency and missing data. The latter are taken into account by the Kalman filter (Kalman, 1960), using a prior which replaces the missing value, i.e., no information leads to no change in distributions for model parameters (West and Harrison, 1997). For other examples of DLM applications, readers are referred to Soudant et al. (1997), Scheuerell et al. (2002), and Hernández-Fariñas et al. (2014, 2017). ~~The time-series were modeled using Dynamic Linear Models (DLM; West and Harrison, 1997) with the dlm package (Petris, 2010) in R software (R core team 2016). This approach is also referred to as State Space Time Series Analysis, Structural Time Series Model, Unobserved Component Model (Harvey et al., 1998) or Dynamic Harmonic Regression (Taylor et al., 2007). This method, associated with the Kalman filter (Kalman, 1960), was also described in Minaudo et al. (2015) and Halliday et al. (2012).~~

The model used was a second order polynomial trend, which allows modelling up to quadratic trend. This was chosen because linear trend (i.e., first order polynomial) was too restrictive and cubic trend (i.e., third order polynomial) might lead to an over fitted model. For the seasonal component, the model used was trigonometric with two harmonics, which allows modelling up to bimodal pattern. This bimodal pattern is characterized by two peaks per year, such as spring and autumn or summer and winter blooms. This model specification was used for all parameters. ~~The model decomposes an observed time-series into component parts, typically trend, seasonal component (i.e., seasonality) and residual as white noise. DLM is~~

~~particularly suitable for environmental data characterized by outliers, irregular sampling frequency and missing data (Hernández-Fariñas et al., 2014). The sequential DLM approach trivially accounts for them: absence of data leads to no change in distributions for model parameters, which look like interpolations. The model used in the current study is a second order polynomial trend, which allows adapting trend up to quadratic, and a two harmonics trigonometric seasonal component, which allow adapting to seasonality up to bimodal.~~

The time unit was defined as the smallest time interval between sampling dates within a period of analysis (i.e., one year). The time unit was weekly, fortnightly or monthly according to sampling frequencies of variables (see Table S1). Normality of standardized residuals was checked using QQ-plot and their independence using estimates of autocorrelation function. If deviations were suspected, outliers were identified as 2.5 % higher and lower than standardized residuals and treated appropriately, i.e., specific observational variances were estimated for each outlier. The DLM time-series analysis provides figures allowing the visual identification of trends and variations in seasonality. ~~The time units based on sampling frequencies were weekly for VB time series, and monthly and bimonthly for the rivers. A variance-stabilizing log transformation was applied to all variables: base was e, except for phytoplankton counts and their ratios, for which the base was 10. Normality of standardized residuals was checked visually using QQ-plot and their independence using estimates of autocorrelation function. If deviations were suspected, outliers were identified as 2.5 % higher and lower standardized residuals and treated appropriately, i.e., specific observational variances were estimated for each of them. For other examples of DLM applications, readers are referred to Soudant et al. (1997), Hernández-Fariñas et al. (2014), and Hernández-Fariñas et al. (2017).~~

2.3.3 Trend

The DLM trend plot displayed observed values with a shade of color for each time unit segments: weekly, fortnightly or monthly. The trend was represented by a dark grey line with the shaded area indicating the 90% confidence interval. For the longest common record of all variables, 1997-2013 called the “common period”, a monotonic linear trend significance test was performed on DLM trend components using a modified non-parametric Mann-Kendall (MK) test (Yue and Wang, 2004). When monotonic linear trends were significant (p<0.05), changes were calculated from differences between the beginning and the end of the common period of the Sen’s robust line (Helsel and Hirsch, 2002).~~For all trend and seasonality figures, the trend plot displayed observed values, with colored dots corresponding to the season. The y-axis represented logarithmic values of studied variables. For a given period, the trend, designated by a dark grey line, was considered as significant if its confidence intervals (shaded area) at the beginning and the end of the period do not overlap. The seasonality plot indicated two aspects: the position of maxima and minima, and the amplitude of seasonal cycles in studied variables. As in the trend plot, the y-axis in the seasonality plot represented the logarithmic values of studied variables. Variations in seasonality can be detected through a change in position of maximum or minimum values within the season if any and/or an increase or a decrease in the seasonal amplitude.~~

2.3.4 Seasonality

The seasonality plot displayed the DLM seasonal component values. The figure gave a visual access to the inter-annual evolution of the amplitude, corresponding to the difference between the minimum and maximum values of each year. As dependent variables have been log-transformed, the model was multiplicative. Therefore, when seasonal component values equaled to 1 (i.e., horizontal line), fitted values equaled to the trend. The seasonality plot also allowed a visualization of how the values have evolved over the years according to their seasonal position. The significance of changes in the seasonality (monotonic linear increase or decrease in the value for a given season) was assessed for the common period using the modified MK test performed on DLM seasonal components for each season. The seasons were defined as winter (January, February, March), spring (April, May, June), summer (July, August, September), and autumn (October, November, December). The interpretation of the seasonal components per se was not meaningful, therefore changes were not calculated, but when monotonic linear trends were significant ($p<0.05$), the sign and the percentage of the changes were provided.~~In order to corroborate the overall trends observed by DLM, a modified non-parametric Mann-Kendall (MK) test (Hamed and Rao, 1998) was performed using deseasonalized observations (i.e. trends) from DLM as an overall annual statistic. The slope of the trend was estimated using Kendall-Theil robust line (Helsel and Hirsch, 2002), a non-parametric approach based on the method of Theil (1950) and Sen (1968). The MK test was also used to corroborate seasonal trends. The seasons were divided as follows: winter (JFM), spring (AMJ), summer (JAS), and autumn (OND). The MK test was applied to the common time segment of all river and Vilaine Bay variables (1997-2013).~~

2.4 Correlation analysis

Spearman Correlations were computed for annual median values of the common period in order to analyze relationships between variables, and tested using STATGRAPHIC CENTURION software (Statgraphics Technologies Inc., Version XVII, Released 2014).~~Spearman Correlations were computed for annual median values in order to analyze relationships among variables, and tested using STATGRAPHIC CENTURION software.~~

3~~.~~ Results

3.1 Long term trends in eutrophication parameters in river basin outlet~~River trends~~

The daily discharge of the Loire varied between ~~157~~ 111 and ~~3,425~~4,760 m^3 s^{-1} ~~from~~ for the period 1980~~ to~~ -2013, with ~~an overall median of 614 m^3 s^{-1}. The~~ DLM trend ~~plot of the Loire discharge~~ display~~ed~~ing oscillations with periodicities of 6-7 years ~~and no noticeable overall trend~~ (Fig. 2a). A significant negative trend was detected for the common period (~~The MK test performed for the period of~~ 1997-2013), with ~~revealed a~~a decrease of 94 m^3 s^{-1} ~~over 16 years~~ (Table 1). The seasonality plot displayed ~~No~~ no marked change, with maximum values always observed in winter (blue) and minimum in summer (orange/red, Fig. 2b) and ~~was observed in the seasonality (Fig. 2b), as confirmed by MK test with~~ no significant linear change ~~trend~~ whatever the season (Table ~~1~~2). The Vilaine discharge, ~~(~~median of ~~36~~ 32 m^3 s^{-1} for the period ~~of~~ 1980-2013), ~~represented~~ corresponded ~~only~~ to 6 % of the Loire discharge and displayed similar trend and seasonality to those of the Loire (Fig. S2, Table 1, 2), as highlighted by the significant correlation between their annual medians (Table 3).

~~Annual median~~ DIP in the Loire varied between ~~decreased from around 2~~0.~~5~~ 1 and 9.4 µmol L^{-1} ~~in the early 1990s to values near 1.5 µmol L^{-1} in 2010s~~for the period 1990-2013 (Fig. 3a~~),~~). A significant decrease of 0.85 µmol L^{-1} ~~which~~ was ~~confirmed by MK test for the period of 1997-2013~~detected for the common period (Table 1). Also during this period, ~~The~~ the seasonality plot indicated a noteworthy shift in ~~position~~ timing of annual DIP ~~minimum~~ minima from summer to spring, as indicated by its change in color from yellow/orange (summer) in 2000 to green (spring) from 2006 onwards ~~observed particularly between 1997 and 2013~~ (Fig. 3b). This change was accompanied by ~~The MK test confirmed this seasonality pattern with~~ a significant negative trend for winter-spring ~~values~~ seasonal components and a significant positive trend for summer-autumn ~~values~~ones (Table ~~1~~2). DIP loads from the Loire ranged between <0.1 and 15 mol s^{-1} for the period 1990 2013, ~~The~~ with trend ~~of DIP loads from the Loire~~ display~~ed~~ing oscillations reflecting the influence of river discharge (Fig

3c). For the common period, the Loire DIP loads ~~with a~~decreased significantly ~~decreasing trend in their annual medians~~by 52% (Table 1). The seasonality plot of DIP loads from the Loire reflected that of discharge with annual minimum and maximum values always observed ~~respectively~~ in summer and winter respectively (Fig. 3d). ~~Patterns~~ Trends of DIP and DIP loads for the Vilaine were similar to those for the Loire (Fig. S3, Table 1, 2), as indicated by a significant correlation between annual medians of DIP in the two rivers (Table 3).

DIN in the Loire ranged between 11 and 489 µmol L^{-1} for the period 1980-2013, with trend ~~displayed~~ displaying a decrease between the 1980s and the early 1990s, followed by an increase~~similar oscillations as those observed for river discharge~~ (Fig. 4a). ~~Annual median DIN decreased from around 150 µmol L^{-1} in the 1980s to the lowest value in the early 1990s (median of 110 µmol L^{-1}) and increased afterwards to reach values around 200 µmol L^{-1}.~~ However, the increase was not significant for the common period ~~The MK test for DIN in the Loire did not indicate a significant trend from 1997 to 2013~~ (Table 1). ~~However, t~~The DLM Loire DIN seasonality plot indicated ~~of DIN in the Loire was marked by~~ a decrease in the seasonal amplitude ~~from the early~~starting in 1990~~s~~ (Fig. 4b~~),~~). For the common period, this decreasing amplitude resulted from ~~with~~ a significant ~~positive trend~~decrease in winter DIN maxima ~~for summer values~~on the one hand and ~~a~~ significant ~~negative trend for winter values~~increase in summer minima on the other hand (Table ~~1~~2) by around 60 µmol L^{-1} (Fig.4a). The DIN loads from the Loire varied from <1.0 to 1,142 mol s^{-1} and displayed similar trend and seasonality to those of DIN (Figs. 4c, 4d~~, Table 1~~)~~.~~, with an increase in summer minima from around 5 to 50 mol s^{-1} for the common period (Fig. 4c, Table 2). The ~~DLM~~ trend of DIN in the Vilaine ~~revealed~~ displayed an oscillation (Fig. S4), with ~~an increase before the mid-1990s and~~ a slight significant decrease over the common period ~~subsequently~~ (~~Fig. S4a~~Table 1) and no marked variation in the seasonality (Fig. S4b, Table 2). ~~The latter was significant according to the MK test (Table 1). The seasonality plot of~~

~~DIN in the Vilaine did not display a marked variation (Fig. S4b).~~ As for the Loire, the trend and seasonality of DIN loads from the Vilaine were similar to those of DIN (Figs. S4c, ~~S4~~d, Table 1, 2).

DIN:DIP ratios in both rivers ranged between 1.0 and 1,000~~,~~ with >80% of value being higher than 30 and displayed an increasing trend between 1990 and 2013 (Fig. S5). A significant increase of 85% and 303%, respectively for the Loire and the Vilaine, was detected for the common period (Table S3). DSi in rivers ranged between 46 and 261 µmol L^{-1} in the Loire and from 5.0 to 201 µmol L^{-1} in the Vilaine for period of available data (2002-2013). More than 80% of DIN:DSi ratios in rivers were higher than the theoretical molar N:Si ratio of 1 for potential requirement of diatoms (data not shown). ~~A marked majority of DIN:DIP ratios in both rivers were above the theoretical molar N:P ratio of 16 for phytoplankton (Redfield, 1958), with a trend increasing from 1990 onwards (Fig. S5; MK, p<0.05). Most of the data of DIN:DSi ratios were higher than the theoretical molar N:Si ratio of 1 for diatoms (Brzezinski, 1985).~~

Chl *a* in the Loire ranged between >200 µg L^{-1} during the 1980s and <1.0 µg L^{-1} in the 2010s. The Chl *a* trend ~~in the Loire was~~remained stable between 1980 and 2000 ~~with annual medians around 60 µg L^{-1} and then~~before ~~decreased~~ decreasing ~~strongly from 2000 to reach median values below 10 µg L^{-1} from 2010 onwards~~subsequently (Fig. 5a). For the common period, ~~A significant~~the Loire Chl *a* decreased ~~was~~ by 93% (54 µg L^{-1}, Table 1).~~also indicated by MK test for the period of 1997-2013 (Table 1).~~ The DLM Loire Chl *a* seasonality plot displayed ~~The decreasing trend in annual Chl *a* in the Loire was accompanied by~~ a shift in ~~position~~ timing of the annual Chl *a* maximum, as indicated by its change in color from orange/red (summer) during 1980-1990 to green (spring) during 2005-~~-~~2013 (Fig. 5b). ~~from late summer before 2000 to spring afterwards (Fig. 5b).~~For the common period, ~~This~~ this change in timing ~~seasonality pattern~~ was ~~confirmed by MK test with~~accompanied by a significant negative trend for autumn seasonal components ~~values~~ and significant positive trend for winter and spring ~~values~~ (Table ~~1~~2). ~~DLM and MK r~~Results for Chl *a* in the Vilaine revealed similar trend and seasonality to those in the Loire (Fig. S6, Table 1, 2), as indicated by a significant correlation between Chl *a*~~.~~ ~~Annual~~ annual medians ~~of Chl *a* and DIP~~ in the two rivers ~~correlated significantly with each other~~ (Table 3).

3.2 Long term trends in eutrophication parameters in the VB~~Vilaine Bay (VB) trends~~

DIP in the VB varied between <0.1 and >1.0 µmol L^{-1} with no noticeable trend (Fig.6a). A significant decrease of 0.05 µmol L^{-1} was detected over the common period (Table 1). The seasonality plot of the VB DIP revealed a change in timing of the minimum values, as indicated by its change in color from yellow/orange (summer) before 2006 to green (spring) afterwards (Fig. 6b). This shift was accompanied by a significant negative linear trend for spring seasonal components and a significant positive trend for summer (Table 2).~~For overall studied period, annual DIP medians in VB were close to 0.5 µmol L^{-1}. The DLM trend seemed to be stable between 1997 and 2013 (Fig. 6a) but the MK test indicated a weak but significant decreasing trend (Table 2). The seasonality plot of DIP in VB revealed a change in position of the minimum values (<0.10 µmol L^{-1}) from early summer to spring by 2005 and an increase in summer values (Fig. 6b). This seasonality pattern was confirmed by the MK test with a significant negative trend for spring values and a significant positive trend for summer values (Table 2).~~

DIN in the VB varied between <1.0 and >200 µmol L^{-1} with trend displaying an oscillation (Fig. 6c). A significant increase of 3.2 µmol L^{-1} was detected for the common period (Table 1). The DLM seasonality indicated that this increase was focused on winter (Fig. 6d, Table 2). Annual DIN medians in the VB were positively correlated with those of discharge from the two rivers (Table 3).~~DIN in VB varied from <1 µmol L^{-1} in summer to 223 µmol L^{-1} in winter 2008, with an overall median of 17 µmol L^{-1} for the period of 1997-2013. Annual DIN medians in VB were positively correlated with those of discharge from~~

~~the two rivers (Table 3). DLM trend of DIN in VB did not display a clear trend (Fig. 6c) nor marked change in seasonality (Fig. 6d) with maximum values in winter and minimum in summer. However, the MK test pointed out a significant increase of 3.2 µmol L^{-1} over the 16 years, focused on winter (Table 2).~~

DSi in the VB varied between <1.0 and 100 µmol L^{-1} without noticeable trend (Fig. 6e). For the common period, a significant increase of 3.6 µmol L^{-1} was detected, which was comparable to that of DIN (Table 1). The seasonality did not indicate any particular change (Fig. 6f, Table 2). Annual DSi medians in the VB were positively correlated with those of the Loire discharge and with the VB DIN (Table 3).

DIN:DIP and DIN:DSi ratios in the VB ranged between <1.0 and 650, and from <0.1 to 44 respectively (Fig. S7). Summer values of DIN:DIP and DIN:DSi ratios were often below theoretical values respectively of 16 and 1 for potential requirements of diatoms (Fig. S7). DSi:DIP ratios in the VB ranged between <5.0 and >100, with >80% of values being above the theoretical value of 16 (Fig. S7). The trends for dissolved inorganic nutrient ratios in the VB displayed a significant increase for the common period (Fig. S7, Table S3).~~Annual DSi medians in VB were positively correlated with those of the Loire discharge and with DIN in VB (Table 3). As for DIN, the maximum DSi value was measured in winter 2008 (91 µmol L^{-1}~~, ~~Fig 6e), but unlike DIN, the minimum values were reached in spring and not in summer (Fig. 6f). The MK test detected a significant increase in annual median values, which was comparable to that of DIN (+3.6 µmol L^{-1} over 16 years) but not attributable to a particular season (Table 2). DIN:DIP, DIN:DSi and DSi:DIP ratios increased significantly in VB (Fig. S7; MK, $p<0.05$).~~

Chl *a* in the VB ranged between 0.1 and 116 µg L^{-1}, with trend displaying an increase (Fig. 7a). For the common period, the VB Chl *a* increased significantly by 126% (2.1 µg L^{-1}, Table 1). The seasonality plot of Chl *a* in the VB displayed a shift in the timing of the annual maximum, indicated by its change in color from green (spring) before 2006 to orange/red (late summer) afterwards (Fig. 7b). This change was accompanied by a significant negative linear trend for spring seasonal components (Table 2). Annual Chl *a* medians in the VB were negatively correlated with those of Chl *a* from both rivers and with DIP in the Vilaine (Table 3).~~Annual Chl *a* medians in VB were negatively correlated with DIP and Chl *a* from both rivers (Table 3). Chl *a* in VB displayed an increasing trend (Fig. 7a), which was confirmed by the MK test (Table 2), with an augmentation of 2.1 µg L^{-1} from 1997 to 2013 corresponding to an increase of 126%. The seasonality plot of Chl *a* in VB indicated a shift in the position of the annual maximum from spring to late summer (Fig. 7b). The MK test on seasons pointed out a significant negative trend for spring values (Table 2).~~

Diatom abundances varied between 200 and 1.3 10^{7} cells L^{-1} for the period 1983-2013, with the DLM trend showing an increase (Fig. 7c). For the common period, diatom abundances increased significantly by 227% (90 10^{3} cells L^{-1}, Table 1). Although diatom abundances continued to peak in spring (Fig. 7d), their seasonality plot indicated a significant increase in summer seasonal components over the common period (Table 2). Dinoflagellate abundances were about ten-fold less than those of diatoms, with values ranging between 40 and 3.4 10^{6} cells L^{-1} over the period 1983-2013. Like diatoms, the DLM trend for dinoflagellate abundances in the VB displayed an increase (Fig. 7d). For the common period, dinoflagellates abundances increased by 8 10^{3} cells L^{-1} (108%, Table 1). However, the DLM seasonality plot indicated that summer seasonal components of dinoflagellate abundances, corresponding to dinoflagellate annual peak, displayed a significant decreasing trend over the common period (Fig. 7f, Table 2).~~The DLM trend of diatom abundances in VB suggested an increasing trend between 1983 and 2013 (Fig. 7c), as confirmed by the MK test performed over the 1997-2013 period (+90,000 Cells L^{-1} over 16 years, Table 2). Although diatoms continued to peak in spring, their seasonality plot indicated that their increase occurred mainly in summer at the expense of the spring period (Fig. 7d). The MK test on seasons denoted a significant increasing~~

~~trend in summer diatom abundances (Table 2). Like diatoms, dinoflagellate abundances increased in VB, as confirmed by the MK test for the period of 1997-2013 (Fig. 7e, Table 2), with an augmentation of 8,000 Cells L^{-1} over the 16 years. However, their seasonality plot (Fig. 7f) and the MK test on seasons (Table 2) pointed out that the summer values, which corresponded to the annual peak, displayed a decreasing trend.~~

~~The DLM trend plot of diatom:dinoflagellate ratios suggested an increase between 1983 and 2013 (Fig. 7g), which was confirmed by the MK test with a significant increasing trend for the period of 1997-2013 (Table 2). The seasonality of diatom:dinoflagellate ratios was marked by an increase in the summer minimum values particularly from 1997, finally reaching the autumn value from 2010 onwards (Fig. 7h). This seasonality pattern was corroborated by the MK test with a significant positive trend in summer diatom:dinoflagellate ratios and a significant negative trend in autumn (Table 2).~~

4. Discussion

The sequence of causes and effects between eutrophication in continental aquatic ecosystems and in those located downstream can be studied by observing trends of eutrophication indicators using the same tool and during the same periods. In the present study, eutrophication trajectories in the downstream VB coastal waters during recent decades were examined, through long-term trends of phytoplankton biomass and nutrient concentrations, in relation to the restoration of the eutrophic Loire and Vilaine Rivers. The DLM analysis provided the opportunity to explore trends and changes in seasonality in a visual manner with figures displaying individual data. The modified non-parametric Mann-Kendall test applied to DLM trend and seasonal components of all variables over common period has permitted corroboration of DLM observations. Overall results demonstrate that upstream recoveries from eutrophication were accompanied by increased eutrophication downstream. The significant reduction in P input relative to N was not enough to mitigate eutrophication all along this river

– coastal marine continuum. More reduction of N input, paying particular attention to diffuse N-sources, is necessary to mitigate eutrophication effectively in the VB coastal waters.

4.1 Eutrophication trajectories at the river basin outlet~~The Loire/Vilaine – Vilaine Bay continuum~~

The decrease in Chl *a* in pre-estuarine stations on the Loire and Vilaine Rivers over the past decades reflects the global diminution in eutrophication in north American and European rivers (Glibert et al., 2011; Romero et al., 2013). This decrease in Chl *a* was also observed in the Upper and Middle Loire (Larroudé et al., 2013; Minaudo et al., 2015). However, the Loire did not retrieve its oligotrophic state of the 1930s (Crouzet, 1983). At the studied stations, the annual Chl *a* peak decreased and shifted from late summer to spring (Figs. 8a, 8b). The parallel decrease of DIP and Chl *a* in the Loire and Vilaine Rivers underlines the role of decreasing P in reducing phytoplankton biomass (Descy et al., 2012; Minaudo et al., 2015), as also found in other river systems, such as the Danube (Istvánovics and Honti, 2012), the Seine (Romero et al., 2013), and some Scandinavian rivers (Grimvall et al., 2014). This decreasing trend of DIP is a result of improved sewage treatment, decreased use of P fertilizers and the removal of P from detergents (Glibert, 2010; Bouraoui and Grizzetti, 2011). However, the decline of Chl *a* in both studied rivers began several years after that of DIP when the latter reached limiting concentrations for phytoplankton, as deduced at Montjean on the Loire by Garnier et al. (2018). The change in timing of the annual DIP minima from summer to spring in the Loire and Vilaine Rivers during last decades of the studied period, concomitant with that of the annual peak of Chl *a*, can be explained by the increasingly early depletion of DIP by phytoplankton (see Floury et al., 2012 for the Loire).

The trend of DIN in studied rivers reveals the general trends observed in other large European rivers, showing a slight decrease, a steady trend or even an increase, depending on the degree of fertilizer application in catchment areas (Bouraoui and Grizzetti, 2011; Romero et al., 2013). The increase in summer Loire DIN since the early 1990s was offset by the decrease in winter values, which is related to the reduction in N point source emissions and N fertilizer application (Poisvert et al., 2016; data from French Ministry of Agriculture, S. Lesaint, pers. comm.). An increase summer DIN of several tens of µmol L^{-1} was also reported in the Middle Loire (Minaudo et al., 2015). This increase in summer DIN is the result of a delayed response due to the long transit time of DIN through soils and aquifers in the Loire catchment (up to 14 years; Bouraoui and Grizzetti, 2011). The decreasing DIN uptake by phytoplankton in the Loire, may have also contributed to the increase in summer DIN (Lair, 2001; Floury et al., 2012). Concerning the Vilaine, the slight decrease in DIN from the early 1990s reflects the decrease in N fertilizer application in the Vilaine catchment (Bouraoui and Grizzetti, 2011; Aquilina et al., 2012), which is facilitated by a relatively short transit time of DIN in the Vilaine watershed (~5-6 yr, Molenat and Gascuel-Odoux, 2002; Aquilina et al., 2012).

DSi data series in both rivers were too short to investigate long-term trends and seasonality, but provided values in order to examine nutrient stoichiometry. Larroudé et al. (2013) observed no significant trend in DSi between 1985 and 2008 in the Middle Loire, as also confirmed at Montjean station by Garnier et al. (2018). The decrease in DIP led to the increasing trend of DIN:DIP ratios, and probably DSi:DIP, in both rivers, as was observed in numerous rivers (Beusen et al., 2016). Based on these trends, the DIP limitation has been thus reinforced in studied rivers during the last decades, and potentially in receiving coastal waters, regardless of the season. ~~Watersheds, rivers and coastal waters located at their outlet, constitute a continuum in which anthropogenic pollutions, generated in watersheds, are transported to coastal zones (Vannote et al., 1980). The sequence of causes and effects between eutrophication in continental aquatic ecosystems and in those located downstream can be studied by observing trends of eutrophication indicators using the same tool and during the same periods. In the present study, the extent of decreasing eutrophication in the Loire and Vilaine Rivers during recent decades was examined in~~

~~relation to the downstream coastal waters of VB through long-term trends of phytoplankton biomass and nutrient concentrations. The DLM test provided the opportunity to explore trends and changes in seasonality in a visual manner with figures displaying individual data. The non-parametric Mann-Kendall test applied to all variables has permitted corroboration of DLM observations. Moreover, the use of Kendall-Theil robust line through the data series has highlighted anthropogenically-driven trends despite large fluctuations caused by climate variation.~~

~~The transfer of nutrients from continents to coastal waters is largely determined by freshwater inputs, the dynamics of which depend largely on precipitation in watersheds. Trends in the Loire and the Vilaine discharges displayed similar oscillations due to variations in precipitation in rivers flowing to the North Sea as reported by Radach and Pätsch (2007). The variability in precipitation may mask long-term changes in water quality induced by human activities (Grimvall et al., 2000). The interpretation of DLM trends depend on series length and starting and end points, and thus may differ from the MK test results. The latter indicate a decrease in discharge for both rivers between 1997 and 2013. This decrease in the Loire discharge was also found in the middle section of the river for the period 1977-2008, resulting essentially from abstraction for irrigation and drinking water (Floury et al., 2012). The strong correlation between Loire and Vilaine discharges underlined the similarities between the two rivers concerning the climate (i.e., precipitation). However, with a tenfold higher discharge than the Vilaine, the Loire is probably the main source of nutrients for the northern Bay of Biscay, with a major role in the eutrophication of coastal waters in south Brittany, including Vilaine Bay (Guillaud et al., 2008; Huret et al., 2013; Ménesguen et al., 2018a). Aside from summer flooding, the closure of the Arzal dam during the low-water periods (Traini et al., 2015), makes the contribution to inputs of freshwater as well as nutrients into VB by this river negligible in summer, in comparison to the Loire.~~

4.2 Eutrophication trajectories in the VB~~Phytoplankton in river and coastal waters~~

In contrast to what happened in rivers, eutrophication in the downstream VB coastal waters has worsened during recent decades, as indicated by significant increase in Chl *a*, also confirmed by the significant augmentation of both diatom and dinoflagellate abundances. The increase in Chl *a* in the VB was accompanied by a shift in its annual peak from spring to summer (Figs. 8c, 8d). This modification in the seasonal course of phytoplankton biomass coincides with the increase in diatom abundances, occurring mainly in summer. The dynamics of phytoplankton in the VB during the last decade of the studied period thus underwent important changes: 1) an increase in biomass, 2) a change in timing of the annual peak from spring to summer, 3) a modification in seasonal course of diatoms and dinoflagellates.~~The decrease in Chl a in both the Loire and Vilaine Rivers over the past decades was also observed in the Upper and Middle Loire (Larroudé et al., 2013; Minaudo et al., 2015), reflecting the general reduction in eutrophication in north American and European rivers (Glibert et al., 2011; Romero et al., 2013). In both rivers, the seasonal peak decreased and shifted from late summer to spring (Figs. 8a, 8b). In contrast, Chl a in VB increased between the mid-1990s and the 2000s. Chl a seasonality in VB also revealed a reverse change from those observed in rivers with the seasonal peak shifting from spring to summer (Figs. 8c, 8d). This modification in the seasonal course of phytoplankton biomass in VB was largely due to the increase in summer diatom abundances to the detriment of spring abundances. Unlike diatoms, the increase in dinoflagellate did not focus in summer. Consequently, the summer minimum values of diatom:dinoflagellate ratios increased in recent decades, indicating changes in the seasonal course of diatoms and dinoflagellates. The dynamics of phytoplankton in the VB has then undergone important changes, as shown by an increase in biomass, a change in position of the annual peak from spring to summer, and an increase in diatom:dinoflagellate ratios with a modification in its seasonal course. Seasonal changes in phytoplankton biomass peaks have been reported in many aquatic ecosystems and mostly attributed to climate change induced temperature (see Edwards~~

~~and Richardson, 2004; Racault et al., 2017). However, variations in nutrient loads can also induce a change in the seasonal pattern of phytoplankton biomass and affect the community structure (Feuchtmayr et al., 2012).~~

4.2.1 Increased Chl *a*~~4.3 Nutrient trends in rivers~~

The increase in phytoplankton biomass could result from several causes, namely overfishing, decrease in commercially grown suspension-feeders, increase in temperature, and increase in nutrient inputs. Increased predation on planktonic herbivores could reduce grazing on phytoplankton (Caddy, 2000). In the VB, commercial fishing is banned in order to protect its ecological function as nursery for demersal fish (Désaunay et al., 2006). The decline in fisheries in the Bay of Biscay since the 1990s (Rochet et al., 2005; Lassalle et al., 2012) was unlikely to have caused increased Chl *a* in the VB, since phytoplankton biomass in these oceanic waters has always been lower than that in the VB (Table S2). Grazing activity by bivalve suspension-feeders can modify phytoplankton biomass (Cloern, 1982; Souchu et al., 2001). In the VB, there was an increase in commercial mussel production (*Mytilus edulis*) between 2001 and 2012 (Le Bihan et al., 2013). This should have led to depletion in phytoplankton biomass, in fact the opposite trend was observed. In regions where the phytoplankton productivity is limited by light availability, an increase in sea surface temperature can promote phytoplankton growth due to water column stabilization (Doney, 2006; Boyce et al., 2010) and decreased turbidity (Cloern et al., 2014). In the VB, except during winter and high hydrodynamic activity periods, phytoplankton production is limited by nutrients (Guillaud et al., 2008). Therefore, the increase in Chl *a* in the VB was particularly due to enhanced nutrient availability, as also reported in China Sea coastal waters by Wang et al. (2018).~~Significant correlations between Chl a and DIP in the Loire and Vilaine Rivers underline the role of decreasing P in reducing phytoplankton biomass (Descy et al., 2012; Minaudo et al., 2015), as also observed in other river systems, such as the Danube (Istvánovics and Honti, 2012), the Seine (Romero et al., 2013), and Scandinavian rivers (Grimvall et al., 2014). The decline of Chl a in the rivers began several years after that of DIP when the~~

~~latter reached limiting concentrations for phytoplankton (Garnier et al., 2018). The change in position of the seasonal DIP minimum from summer to spring in the Loire and Vilaine Rivers during last decades (Figs. 8a, 8b), concomitant with that of the seasonal peak of Chl a, can be explained by the increasingly early depletion of DIP by phytoplankton (see Floury et al., 2012 for the Loire).~~

4.2.2 Changes in timing of annual Chl *a* peak

Seasonal changes in phytoplankton biomass peaks have been reported in other aquatic ecosystems and mostly attributed to climate change-induced temperature (Edwards and Richardson, 2004; Racault et al., 2017). Variations in nutrient availability can also induce a change in the seasonal pattern of phytoplankton biomass (Thackeray et al., 2008; Feuchtmayr et al., 2012). These authors observed that the advancement in the timing of the spring diatom bloom in some English lakes was related to the increase in winter DIP. In the VB, the shift in annual Chl *a* peak from spring to summer, coupled with the change in position of the annual DIP minima from summer to spring, suggests that DIP depletion by phytoplankton bloom occurred progressively earlier during the last two decades. Based on nutrient concentrations and stoichiometry (Justić et al., 1995), the first nutrient limiting phytoplankton biomass in the VB shifts seasonally from DIP in spring to DIN in summer, as verified by bioassays (Retho et al. Ifremer, unpublished data). The conjunction of the decrease in DIP and an increase in DIN in the VB has probably also contributed to the shift in annual Chl *a*.~~The DLM trend of DIN in the Loire suggests an increase in values since the 1990s, which was not confirmed by the MK test over the period of 1997-2013. However, the DLM seasonality and the MK test on seasons suggest that the increase in summer DIN was offset by the decrease in winter values. This increase in summer DIN was also observed in the Middle Loire by Minaudo et al. (2015) and Floury et al. (2012). These authors reported an increase of approximately 50 µmol L-1 during the last decades. This was probably the result of a delayed response due to the long transit time of DIN through soils and aquifers in the Loire watershed (up to 14 years;~~

~~Bouraoui and Grizzetti, 2011), and this in spite of the reduction in N point source emissions and N fertilizer application (Poisvert et al., 2016; data from French Ministry of Agriculture, S. Lesaint, pers. comm.). The decreasing DIN uptake of by phytoplankton in the Loire during past years, may have also contributed to the increase in DIN (Lair, 2001; Floury et al., 2012). DSi data series in both rivers were too short to investigate long-term trends and seasonality. However, Larroudé et al. (2013) observed no significant trend in DSi between 1985 and 2008 in the Middle Loire, as also confirmed by Garnier et al. (2018) at the Montjean station upstream Sainte-Luce (Fig 1). Consequently, during last two decades, VB has received decreasing DIP inputs and increasing DIN inputs, especially from the Loire, but no change in those of DSi, in accordance with trends in concentrations. Therefore, the change in stoichiometry of nutrient loads into VB coastal waters is likely considering the increase in N:P (and N:Si) ratios, as also pointed out by Grizzetti et al. (2012) for most large rivers on Atlantic coasts. (Brzezinski, 1985).~~

4.2.3 Role of DSi on seasonal course of diatoms and dinoflagellates

In terms of nutrients, the balance between diatoms and dinoflagellates is predominantly regulated by the DSi availability (Egge and Aksnes, 1992). In the VB, based on nutrient concentrations and stoichiometry, diatoms were rarely limited by the DSi availability, thanks probably to internal DSi regeneration, as suggested by Lunven et al. (2005) and Loyer et al. (2006) in the northern Bay of Biscay continental shelf. The fact that diatoms have increased more than dinoflagellates in the VB, contradicts the idea that excessive DIN and DIP inputs favor phytoplankton species, which do not require DSi (Conley et al., 1993; European Communities, 2009; Howarth et al., 2011). An increase in diatom abundances during the eutrophication process was also observed in Tolo Harbor (Yung et al., 1997; Lie et al., 2011) and the coastal waters of the Gulf of Finland (Weckström et al., 2007). Conversely, decreasing eutrophication in the Seto Inland Sea (Yamamoto, 2003), in Thau (Collos et al., 2009) and other Mediterranean Lagoons (Leruste et al., 2016) was accompanied by the increase in dinoflagellate abundances to the detriment of diatoms. These observations and our results provide evidence that eutrophication can be manifested by an increase in diatom abundances.

~~4.4 Nutrient trends in coastal waters~~4.3 Loire/Vilaine - VB continuum

In theory, several external nutrient sources could have contributed to nutrient availability in the VB: atmospheric, oceanic and riverine inputs. DIN inputs from rainwater estimated by Collos et al. (1989) represent only 1% of river inputs, while levels of nutrients and Chl *a* in the Bay of Biscay always remained low during the studied period (Table S2). The proximity of the VB to the Loire and Vilaine Rivers designates riverine inputs as main external nutrient sources in these coastal waters (Ménesguen et al., 2018a, b).

4.3.1 Rivers as the main external nutrient source to the VB

Watersheds, rivers and coastal waters located at their outlet, constitute a continuum in which anthropogenic pollution, generated in watersheds, are transported to coastal zones (Vannote et al., 1980; Bouwman et al., 2013). The transfer of nutrients from continents to coastal waters is largely determined by freshwater inputs, the dynamics of which depend largely on precipitation in watersheds. Trends in the Loire and the Vilaine discharges displayed similar oscillations to those of rivers flowing to the North Sea as reported by Radach and Pätsch (2007), suggesting a common hydro-climatic pattern in Western Europe linked to the North Atlantic Oscillation. The decrease in the Loire discharge observed between 1997 and 2013 was also found in the middle section of the river for the period 1977-2008 (Floury et al., 2012) and attributed essentially to abstraction for irrigation and drinking water by these authors. The strong correlation between Loire and Vilaine discharges underlines the similarities between the two rivers concerning the precipitation regime. However, with a tenfold higher discharge than the Vilaine, the Loire remains the main source of freshwater for the northern Bay of Biscay, with a major role in the eutrophication of coastal waters in south Brittany, including the VB (Guillaud et al., 2008; Ménesguen et al., 2018a, 2019). Aside from flood periods, the closure of the Arzal dam during the low-water periods (Traini et al., 2015), makes nutrient inputs into the VB by the Vilaine negligible in summer, compared to those from the Loire.~~Among the different drivers of change in the phytoplankton biomass, namely temperature, fishing and nutrient inputs, the latter are probably the main cause of the modifications observed in VB. In theory, several external nutrient sources could have contributed to the increasing trend of Chl a in VB: atmospheric, oceanic and fluvial inputs. DIN inputs from rainwater estimated by Collos et al. (1989) represent only 1% of river inputs, while levels of nutrients and Chl a the Bay of Biscay always remained low during the study period (Table S2). Significant negative correlations between Chl a in VB, and Chl a and DIP in rivers, as well as significant positive correlations between DIN and DSi in VB and river discharge, suggest that the change in eutrophication parameters in VB (i.e., phytoplankton biomass) was directly related to changes in rivers. Moreover, increasing DIN:DIP and DSi:DIP ratios in VB mirrors those observed in rivers. Consequently, the dynamic of phytoplankton in VB must be interpreted essentially according to variations in riverine nutrient inputs, especially those from the Loire (Ménesguen et al., 2018a, b).~~

4.3.2 Role of estuaries and the Vilaine dam

Biogeochemical processes within estuaries may alter the nutrient transfer from rivers to coastal waters (Statham, 2012; Jickells et al., 2014). Coupled nitrification-denitrification and ammonification-anammox can be a sink of N in estuaries (Howarth et al., 1996; Abril et al., 2000). Inorganic nutrients in estuaries can also be removed by phytoplankton uptake, which is nonetheless limited by turbidity (Middelburg and Nieuwenhuize, 2000; Guillaud et al., 2008). Estuaries can also act as a source of nutrients, resulting from mineralization of riverine phytoplankton organic matter (Meybeck et al., 1988; Middelburg et al., 1996; Etcheber et al., 2007). However, for the studied rivers, this process may have diminished with the decreasing trend in riverine Chl *a*. The desorption of loosely bound P from suspended mineral particles on arrival in saline waters can also provide a source of DIP (Deborde et al., 2007; van der Zee et al., 2007). Except during flood periods, the suspended particle fluxes from the Loire are generally low (Moatar and Dupont, 2016). In addition to these biogeochemical processes, the increase in population around the Loire estuary (ca. 1% per year, INSEE, 2009) during the last decades may have contributed to the increase in N and P inputs. However, inputs of DIN and DIP from wastewater treatment plants in the Loire and Vilaine estuaries have not increased due to improved treatment techniques (Loire-Brittany River Basin Authority, P. Fera, pers. comm.). The presence of a dam at the river outlet may increase water residence time, thus favoring nutrient uptake by phytoplankton and loss of N via denitrification (Howarth et al., 1996; Seitzinger et al., 2006). Unfortunately, for these two studied rivers, processes in estuaries and dam are poorly investigated and quantified, which makes it difficult to estimate their influence on nutrient transfer to coastal zone.

Despite influences of estuaries and dam, the increase in DIN:DIP and DSi:DIP ratios in rivers during last two decades, with values already largely above the theoretical value of 16 in the 1990s, has been reflected in the VB coastal waters (Figs. S5, S7). Although biogeochemical processes in estuaries and the Vilaine dam may introduce bias in nutrient transfer from rivers to the VB, they are probably not intense enough to decouple the observed trends between rivers and the VB, as suggested by Romero et al. (2016) for the Seine River – Seine Bay continuum. Moreover, significant negative correlations between annual Chl *a* medians in the VB and in rivers, as well as significant positive correlations between annual medians of DIN and DSi in the VB with those of river discharge suggest that changes in eutrophication parameters in the VB (i.e., phytoplankton biomass) were related to changes in rivers (Ménesguen et al., 2018a, b).~~In VB, the first nutrient limiting phytoplankton biomass shifts from DIP in spring to DIN in summer (Retho et al. unpublished data). This seasonal pattern is also observed in other anthropogenically influenced coastal ecosystems, as in Chesapeake Bay (Kemp et al., 2005) and~~

~~northern Gulf of Mexico (Rabalais et al., 2002). The decrease in DIP loads from rivers during the last decades was probably the cause of the simultaneously decreasing trend in the VB and may have reinforced spring DIP limitation as also reported by Billen et al. (2007) in Seine Bay. This result is consistent with the MK decreasing trend of spring Chl a. The worsening eutrophication, which developed rather in summer, was the consequence of increasing DIN inputs from the Loire. Therefore, it is likely that the conjunction of decreasing DIP loads and increasing summer DIN loads from the Loire have generated the seasonal change in annual Chl a peak in VB.~~

4.3.3 Link between eutrophication trajectories in rivers and in the VB

During the last two decades, the downstream VB coastal waters have received decreasing DIP inputs, increasing DIN inputs especially from the Loire during summer, and no change in DSi inputs (Fig. 8). The decrease in riverine DIP loads was the cause of the simultaneously decreasing trend in the VB DIP and may have reinforced spring DIP limitation as also reported by Billen et al. (2007) in the Seine Bay. The worsening eutrophication in the VB was the consequence of increasing DIN inputs from the Loire. A similar observation was reported in other coastal ecosystems, such as the Neuse River estuary (Paerl et al., 2004), Belgian coastal waters (Lancelot et al., 2007), and the Seine Bay (Romero et al., 2013), where decreasing upstream Chl *a*, due to DIP input reduction, was accompanied by the increase in downstream Chl *a*, as a result of increasing DIN input. The seasonal change in annual Chl *a* peak in the VB resulted also from the conjunction of decreasing DIP loads and increasing summer DIN loads from the Loire. The summer limitation of phytoplankton production by DIN in the VB cannot be explained by the stoichiometry of nutrients in rivers. Internal sources of nutrients, especially sediments (see below), were also likely to support a significant portion of nutrient availability for phytoplankton production during the period of low river discharge (Cowan and Boynton, 1996; Pitkänen et al., 2001).

4.3.4 Role of internal nutrient loads

In shallow ecosystems, internal nutrient recycling can regulate phytoplankton production and potentially exacerbate eutrophication (Paerl et al., 2016), as observed both in lakes (Jeppesen et al., 2005) and coastal ecosystems (Pitkänen et al., 2001). Compared to freshwater, the fragility of marine ecosystems is related to salinity (Blomqvist et al., 2004). The presence of sulfate a major element of salinity) decreases the efficiency of sediments to retain DIP (Caraco et al., 1990; Lehtoranta et al., 2009) and favors the recycling of DIP over DIN, the latter being potentially eliminated through denitrification (Conley, 2000; Conley et al., 2009). In the VB, measurements of benthic nutrient fluxes confirm that sediments represent a substantial DIP and DSi source compared to riverine inputs (Ratmaya, 2018), allowing summer phytoplankton production to benefit from surplus DIN inputs from the Loire. Sediments were then able to support phytoplankton production by providing DIP and DSi, as found in other coastal ecosystems (Cowan and Boynton, 1996; Boynton et al., 2008), and probably to switch the first limiting nutrient from DIP in spring to DIN in summer, as observed in the Baltic Sea (Conley, 2000; Pitkänen et al., 2001). Consequently, the increase in summer diatom abundances in the VB was mainly due to increased summer DIN loads from the Loire, sustained by internal sources of DIP and DSi coming from sediments.~~Regarding the trends in nutrient loads from the Loire: increase in DIN, decrease in DIP and stability in DSi (Larroudé et al., 2013), as well was nutrient stoichiometry in rivers, additional sources of DIP and DSi in VB have been necessary for diatoms to benefit from increased summer riverine DIN inputs. The fragility of marine ecosystems is related to the elevated salinity (Blomqvist et al., 2004), which favors the recycling of DIP over DIN in sediments (Caraco et al., 1990; Conley, 2000; Conley et al., 2009). The measurements of benthic nutrient fluxes in summer 2015 provide preliminary evidence of the role played by sediments in the worsening of eutrophication in the VB (Table 4). Benthic DIP and DSI inputs were approximately ten and fourfold higher than those coming from the Loire and Vilaine Rivers. Sediments were then able~~

~~to support summer phytoplankton production by providing DIP and DSi, as found in other coastal ecosystems (Cowan and Boynton, 1996; Boynton et al., 2008), and probably to switch the first limiting nutrient from DIP in spring to DIN in summer, as observed in the Baltic Sea (Conley, 2000; Pitkänen et al., 2001; Bonaglia et al., 2014). Consequently, the increase in summer phytoplankton biomass in VB was mainly due to increased summer DIN loads from the Loire, sustained by internal sources of DIP and DSi coming from sediments.~~

4.~~5~~ 4 Implications for nutrient management

4.4.1 Impact of nutrient management strategies

The need to control both N and P inputs to mitigate eutrophication along the freshwater-marine continuum is still debated within the scientific community (see Schindler et al., 2008; Conley et al., 2009; Schindler, 2012; Paerl et al., 2016; Schindler et al., 2016). Despite the imbalance between P and N input reduction, eutrophication in the river section of the Loire/Vilaine – VB continuum has diminished but the increase in phytoplankton biomass in the VB provides evidence that significant reduction of P inputs, without concomitant N abatement, was not yet sufficient to improve water quality along the entire continuum. Targeting N and P pollution from point sources has successfully reduced eutrophication in marine ecosystems, as evidenced in Tampa Bay (Greening and Janicki, 2006) and in several French Mediterranean lagoons (Collos et al., 2009; Leruste et al., 2016). However, N pollution in coastal waters from rivers with watersheds largely occupied by intensive agriculture remain problematic in many European countries (Bouraoui and Grizzetti, 2011; Romero et al., 2013). Reducing diffuse N inputs through improved agricultural practices and structural changes in the agro-food system (Desmit et al., 2018; Garnier et al., 2018) would probably help to lessen eutrophication (Conley et al., 2009; Paerl, 2009). Assuming that rapid and radical change in farming practices is implemented, the delayed responses due to variations in transit time of NO_3^- in aquifers should be taken into account for restauration strategy (Bouraoui and Grizzetti, 2011).

In the VB, a reduction in DIN inputs especially during the summer would probably have prevented eutrophication from worsening in this ecosystem. Given that in many other coastal ecosystems the first nutrient limiting phytoplankton production tends to switch from DIP in spring to DIN in summer (Fisher et al., 1992; Del Amo et al., 1997; Conley, 2000; Tamminen and Andersen, 2007), it would be relevant to take into account seasonal aspects for nutrient reduction strategy. ~~The Loire and Vilaine Rivers displayed a trend similar to northern European rivers (Lacroix et al., 2007) and the Neuse (Paerl et al., 2004), for which more effective P reduction relative to N did not restore their downstream coastal waters from eutrophication. The long-term trajectories of this case study provide a more evidence that reduction of P inputs alone was not sufficient yet to control eutrophication (i.e., increased phytoplankton biomass) in coastal waters of VB. The need to control both N and P inputs to mitigate eutrophication in coastal waters remains a debate among scientific (see Schindler et al., 2008; Conley et al., 2009; Schindler, 2012). However, dual nutrient reduction strategy implementing actions on wastewater treatment plants (i.e., point sources) is efficient in reducing eutrophication in marine ecosystems, as evidenced in Tampa Bay (Greening and Janicki, 2006), and in several French Mediterranean lagoons (Collos et al., 2009; Leruste et al., 2016). In contrast, reducing N losses in coastal waters influenced by rivers with watersheds largely occupied by intensive agriculture (i.e., diffuse sources) remain problematic (Bouraoui and Grizzetti, 2011; Sebilo et al., 2013). The reduction in diffuse N sources, through better agricultural practices and structural changes in the agro-food system (see Desmit et al., 2018; Garnier et al., 2018), provide options to decrease N losses to coastal waters, thus will probably help to lessen eutrophication (Conley et al., 2009; Paerl, 2009).~~

4.4.2 Influence of internal nutrient regeneration

In the VB, the internal nutrient recycling from sediments appears to have contributed to the worsening of summer water quality during the last two decades and augmented the effects of anthropogenic nutrient inputs. Internal nutrient loads can delay ecosystem recovery from eutrophication following external nutrient input reduction (Duarte et al., 2009). In lakes, this delay induced by internal loads of P on the oligotrophication process varies from 10 to 20 years (Jeppesen et al., 2005; Søndergaard et al., 2007). In coastal ecosystems, the delay resulting from internal nutrient loads was less studied. However, Soetaert and Middelburg (2009), using a model in a shallow coastal ecosystem, estimated a delay of more than 20 years following the reduction of external N input. Therefore, for the Loire/Vilaine – VB continuum, nutrient management strategies should consider the internal nutrient loads in order to anticipate the delay in recovery of the VB coastal waters from eutrophication.

5. Conclusions and perspectives

Parallel investigation of eutrophication parameters in the Loire and Vilaine Rivers, and coastal waters under their influence revealed several striking patterns and relationships, of which the most apparent was upstream recoveries from eutrophication accompanied by increased eutrophication downstream (Fig. 8). During the last two decades, Loire-Vilaine coastal waters have experienced a diminution in DIP inputs, whereas DIN continued to increase in the Loire during summer. While the decreasing trends in DIP were accompanied by declining phytoplankton biomass in rivers, the seasonal cycle of phytoplankton has been changed in downstream VB, with an increase in biomass, a shift in its annual peak from spring to summer, and a modification in the seasonal course of diatoms and dinoflagellates. Moreover, the concept of diatom replacement by dinoflagellates during the eutrophication process does not seem to be applicable to all shallow coastal ecosystems.

These results open up a whole field of investigation into the effects of changes in the phytoplankton dynamics on food webs, which is of major importance to this flatfish nursery and commercial shellfish area (Désaunay et al., 2006; Chaalali et al., 2017). Further studies are necessary to investigate the modifications in the phytoplankton community, especially the phenology of the different species, as well as the possible consequence on food webs. Finally, the internal loads of nutrients from sediments are suspected of counteracting the reduction of external nutrients, thus delaying the restauration progress. During the eutrophication process, sediments may also play an important role in the balance between diatoms and others classes of phytoplankton. Taking into account these internal processes in modelling studies (i.e., ECO-MARS3D, Ménesguen et al., 2018a, b; Ménesguen and Lacroix, 2018), will better simulate nutrient load scenarios in shallow coastal bays (work in progress).~~Parallel investigation of eutrophication parameters in rivers and coastal waters under their influence revealed several striking patterns and relationships, of which the most apparent was upstream recoveries from eutrophication accompanied by increased eutrophication downstream (Fig. 8). During the last two decades, Loire-Vilaine coastal waters have experienced a diminution in DIP inputs, whereas DIN continued to increase in the Loire. While the decreasing trends in DIP were accompanied by declining phytoplankton biomass in rivers, the seasonal cycle of phytoplankton has been changed in downstream VB, with an increase in biomass, a shift in its annual peak from spring to summer, and a modification in seasonal course of diatoms and dinoflagellates. These results open up a whole field of investigation into the effects of changes in the phytoplankton dynamics on food webs, which is of major importance to this flatfish nursery and commercial shellfish area (Désaunay et al., 2006; Chaalali et al., 2017). Further studies are necessary to investigate the modifications in the phytoplankton community, especially on the phenology of the different species, as well as the possible consequence on food webs.~~

Data availability

All data used in this study are available in the following online data bases: French National Observation Network for Phytoplankton and Hydrology in coastal waters (https://doi.org/10.17882/47248), French Oceanographic Cruises PELGAS surveys (http://campagnes.flotteoceanographique.fr/series/18/), Loire-Brittany River Basin Authority (http://osur.eau-loire-bretagne.fr/exportosur/Accueil), French hydrologic database (http://www.hydro.eaufrance.fr/), ICES Oceanographic database (http://ocean.ices.dk/HydChem/HydChem.aspx?plot=yes).

Author contribution

PS and WR designed the study. WR compiled and prepared the datasets. DS performed statistical and time series analyses. WR wrote the manuscript with contributions from all co-authors (PS, DS, JSM, NCL, EG, FAL, LB). Author abbreviations: WR = Widya Ratmaya, DS = Dominique Soudant, JSM = Jordy Salmon-Monviola, NCL = Nathalie Conchennec-Laureau, EG = Evelyne Goubert, FAL = Françoise Andrieux-Loyer, LB = Laurent Barillé, PS = Philippe Souchu.

Competing interests

The authors declare that they have no conflict of interest.

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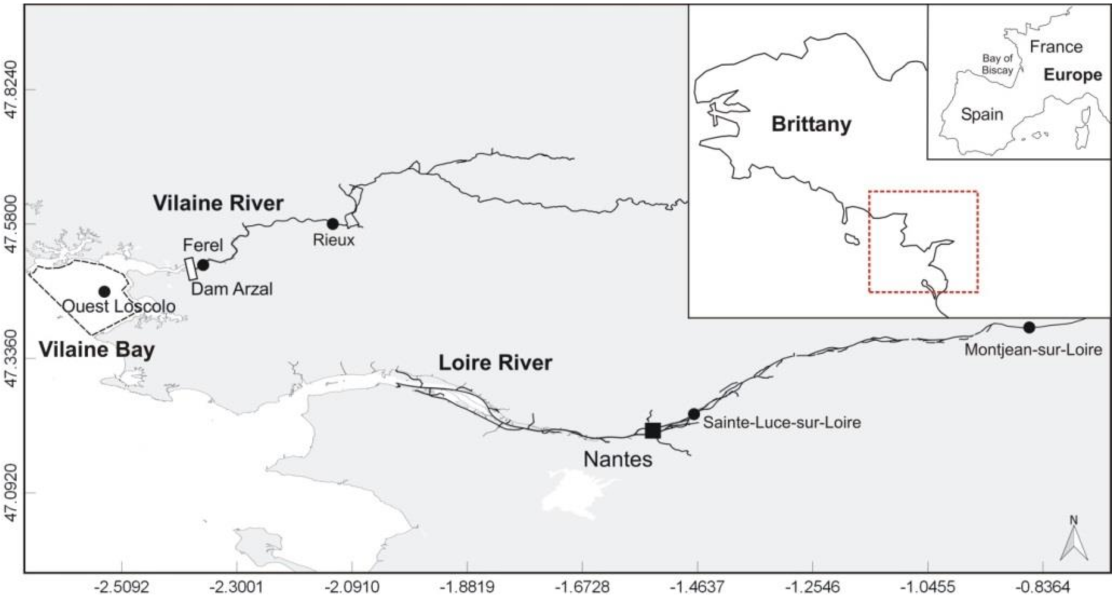
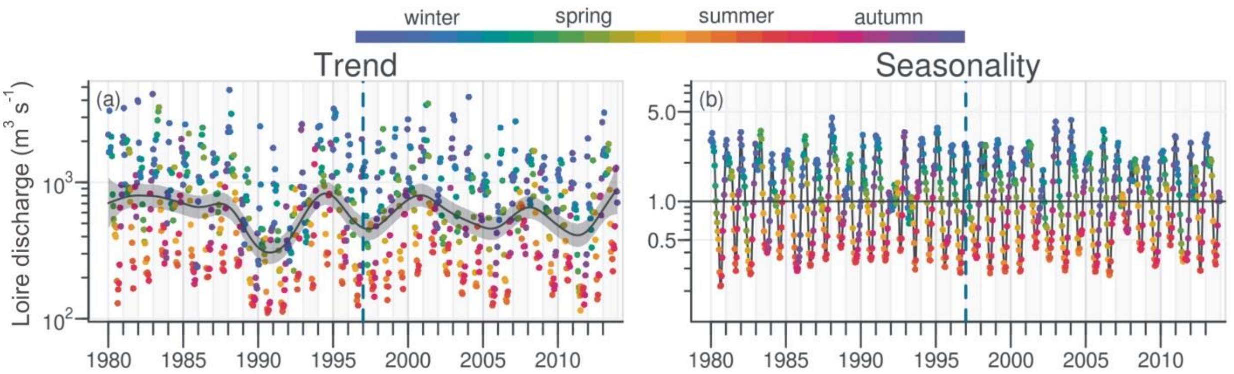


Figure 1: Map of the area studied showing Loire and Vilaine rivers and delimitation of Vilaine Bay (inset red dotted line). Black dots mark the sampling and gauging stations cited



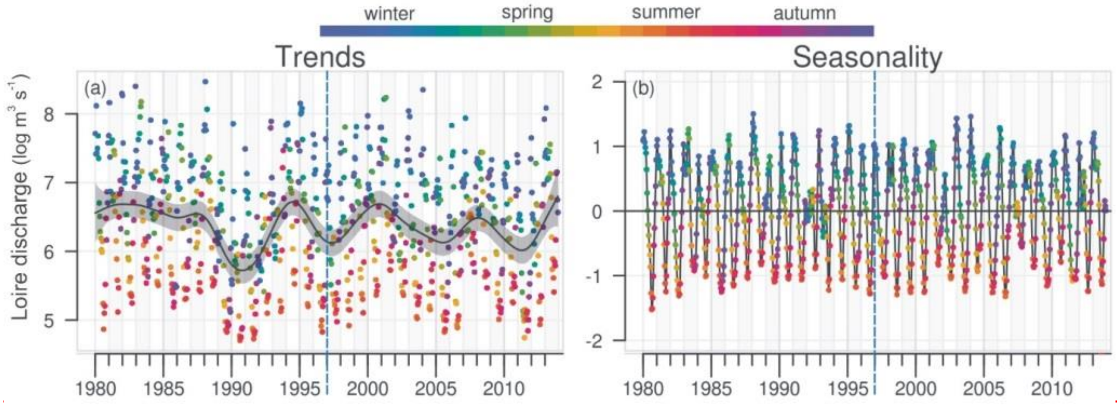
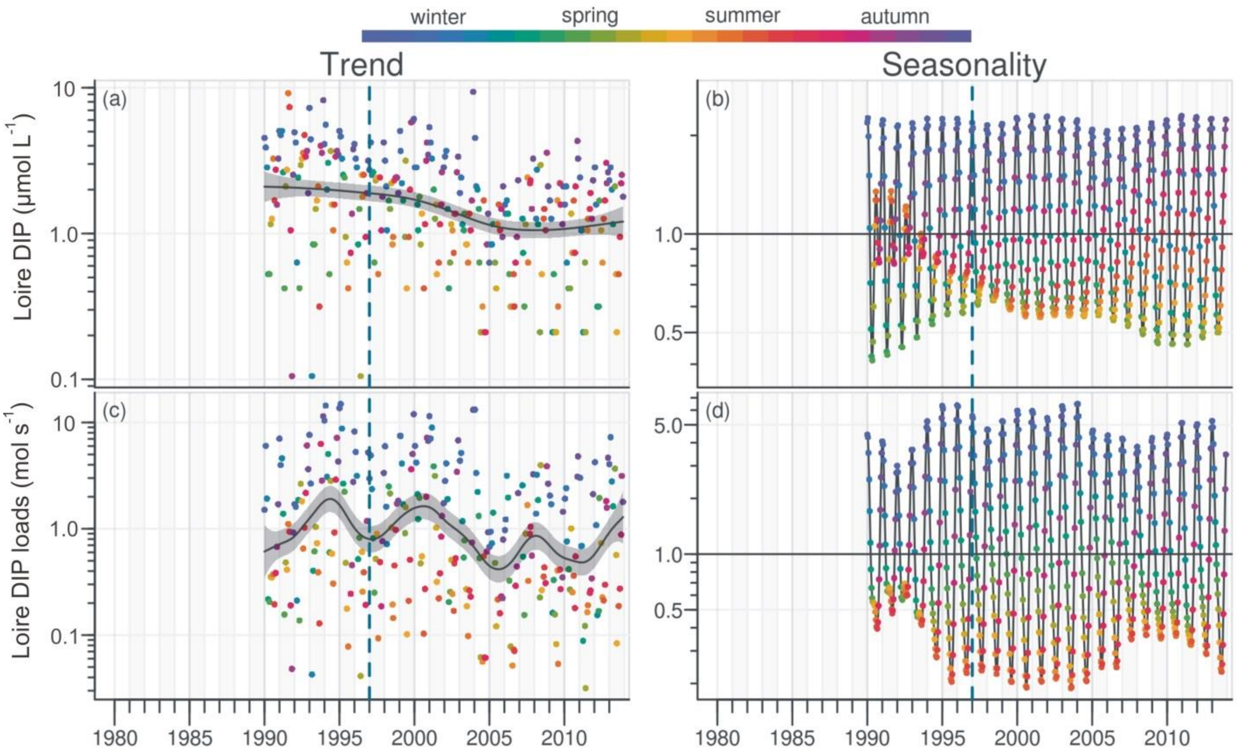


Figure 2: Long-term trend~~s~~ and seasonality of the Loire discharges (a, b). Dark grey lines represent DLM trends. Shaded areas indicate the ~~95~~ 90 % confidence interval. Each dot in the trend plot (left) represents an observed value, those in the seasonality plot (right) represent values estimated by the model. On the seasonality plot, the horizontal line (y = 1.0) indicates seasonal components for which fitted values equal to the trend. Dashed vertical blue line corresponds to the longest common period for all studied variables in rivers and in the VB ~~MK test (1997-2013)~~


winter
spring
summer
autumn
Trend
Seasonality
Loire DIP (µmol L-1)
Loire DIP loads (mol s-1)
(a)
(b)
(c)
(d)
10
1.0
0.1
0.5
5.0
1980
1985
1990
1995
2000
2005
2010

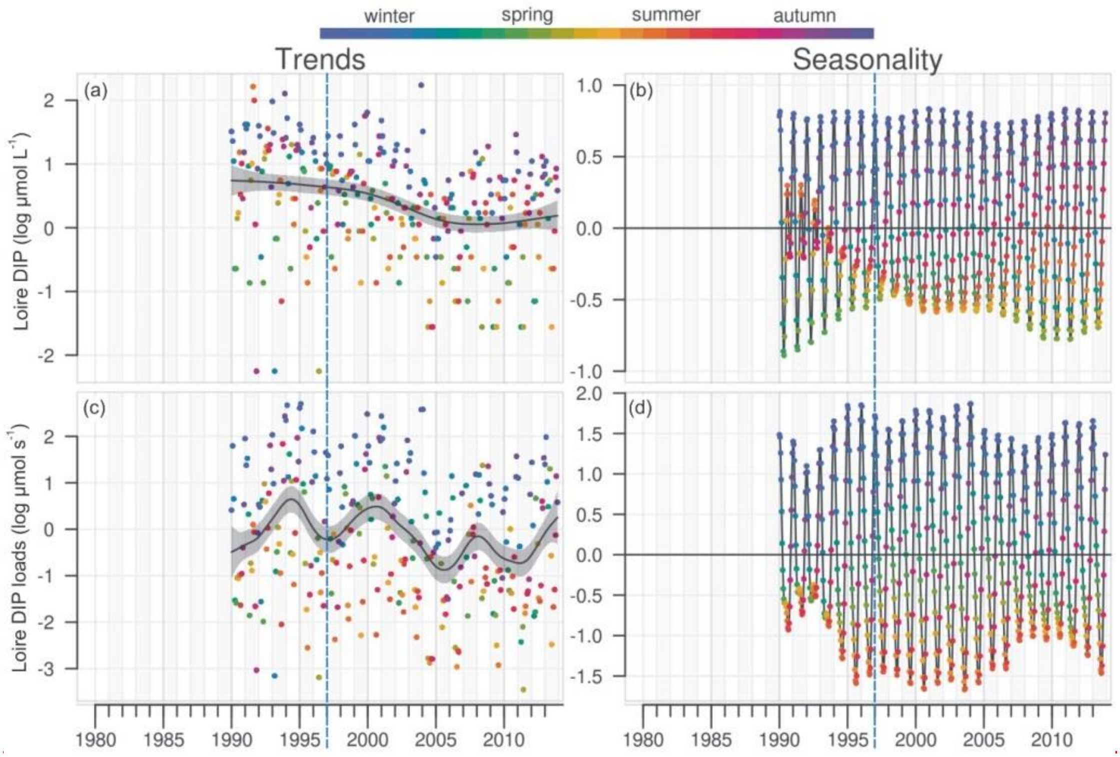
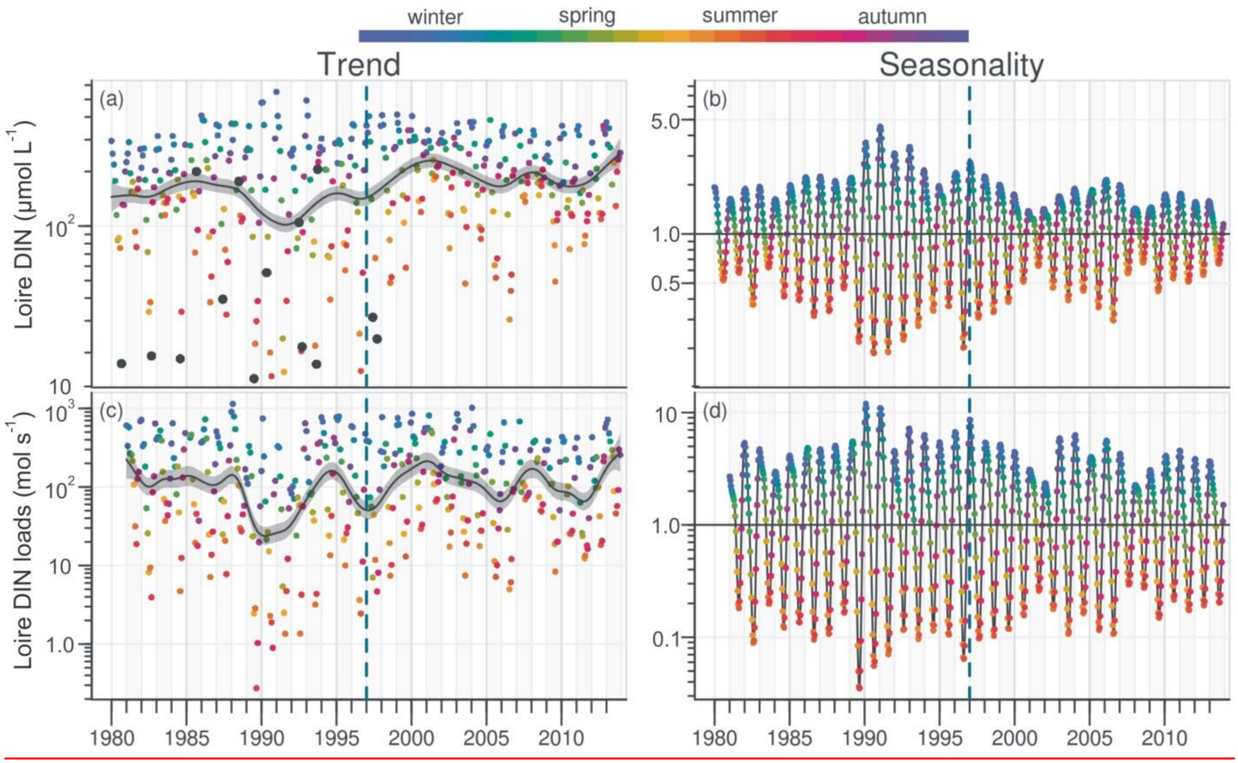


Figure 3: Long-term trend~~s~~ and seasonality of DIP in the Loire (a, b) and DIP loads from the Loire (c, d). ~~Note: analytical problems of DIP measurements before 1990 (see Section 2.4.).~~ See Fig. 2 for details


winter
spring
summer
autumn
Trend
Seasonality
(a)
(b)
(c)
(d)
Loire DIN (µmol L⁻¹)
Loire DIN loads (mol s⁻¹)
10
10²
10³
10
1.0
0.5
1.0
5.0
0.1
1.0
10
1980
1985
1990
1995
2000
2005
2010

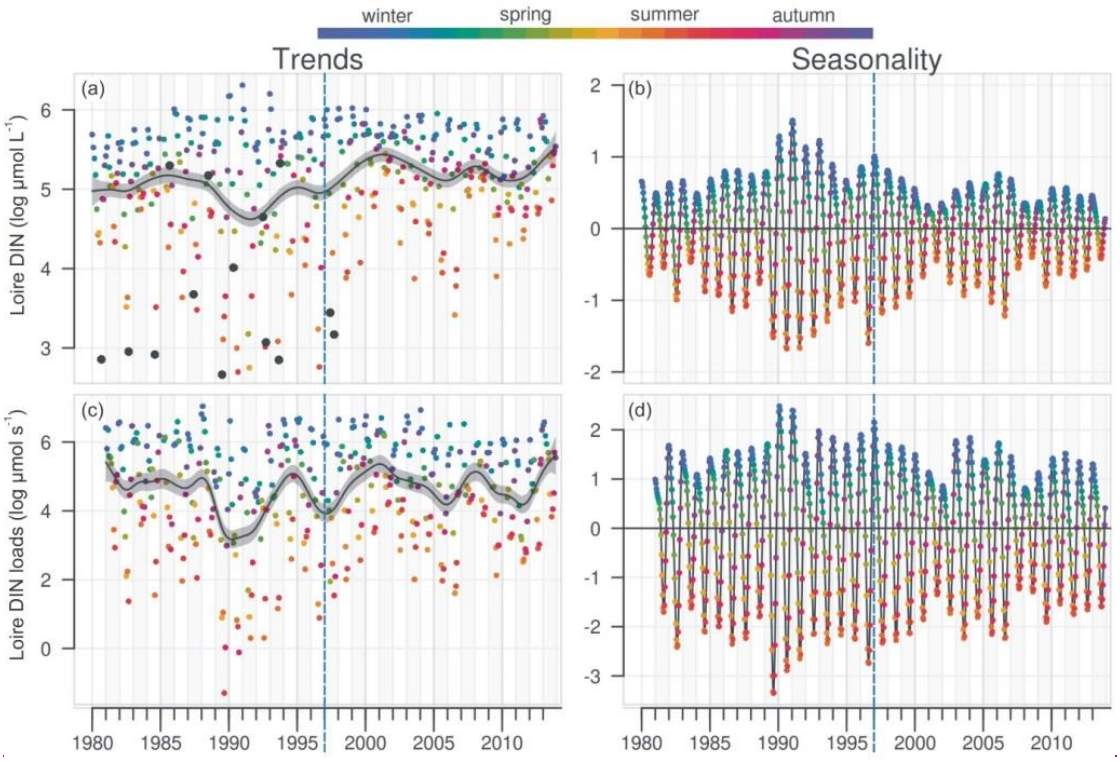
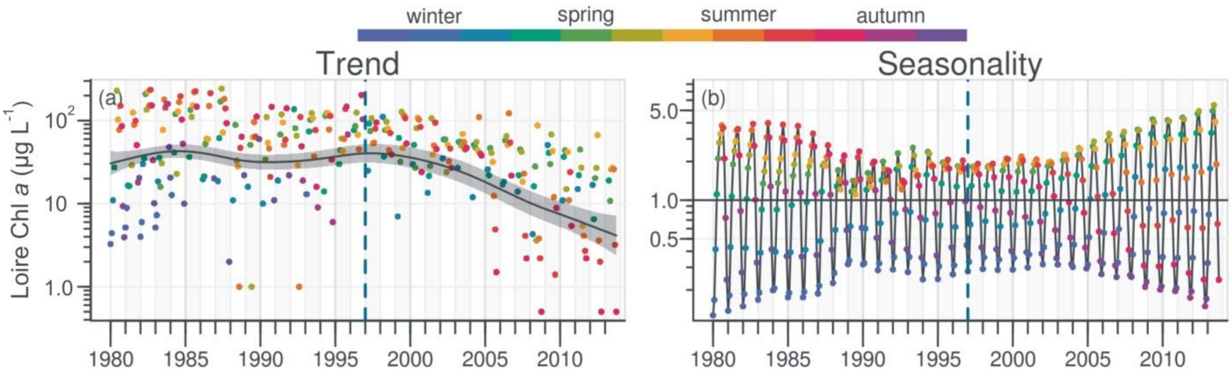


Figure 4: Long-term trend~~s~~ and seasonality of DIN in the Loire (a, b) and DIN loads from the Loire (c, d). Black dots represent data considered as outliers (see Section 2.4.). See Fig. 2 for details



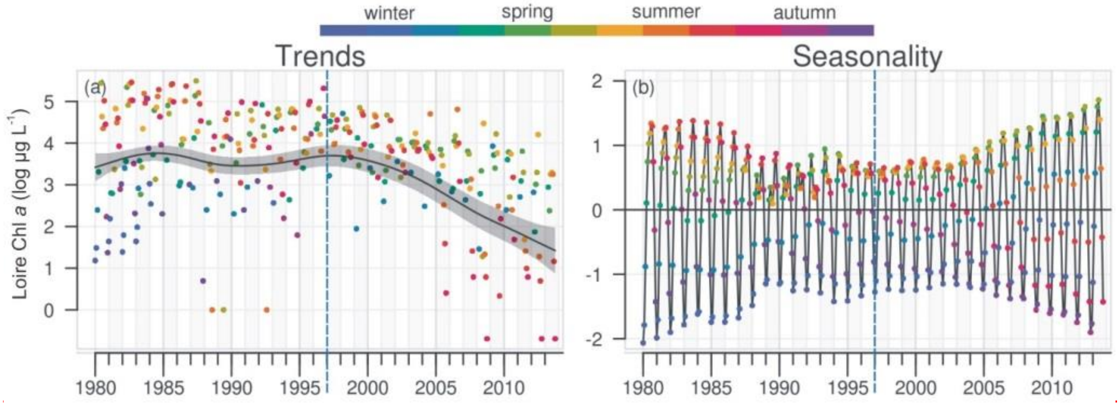
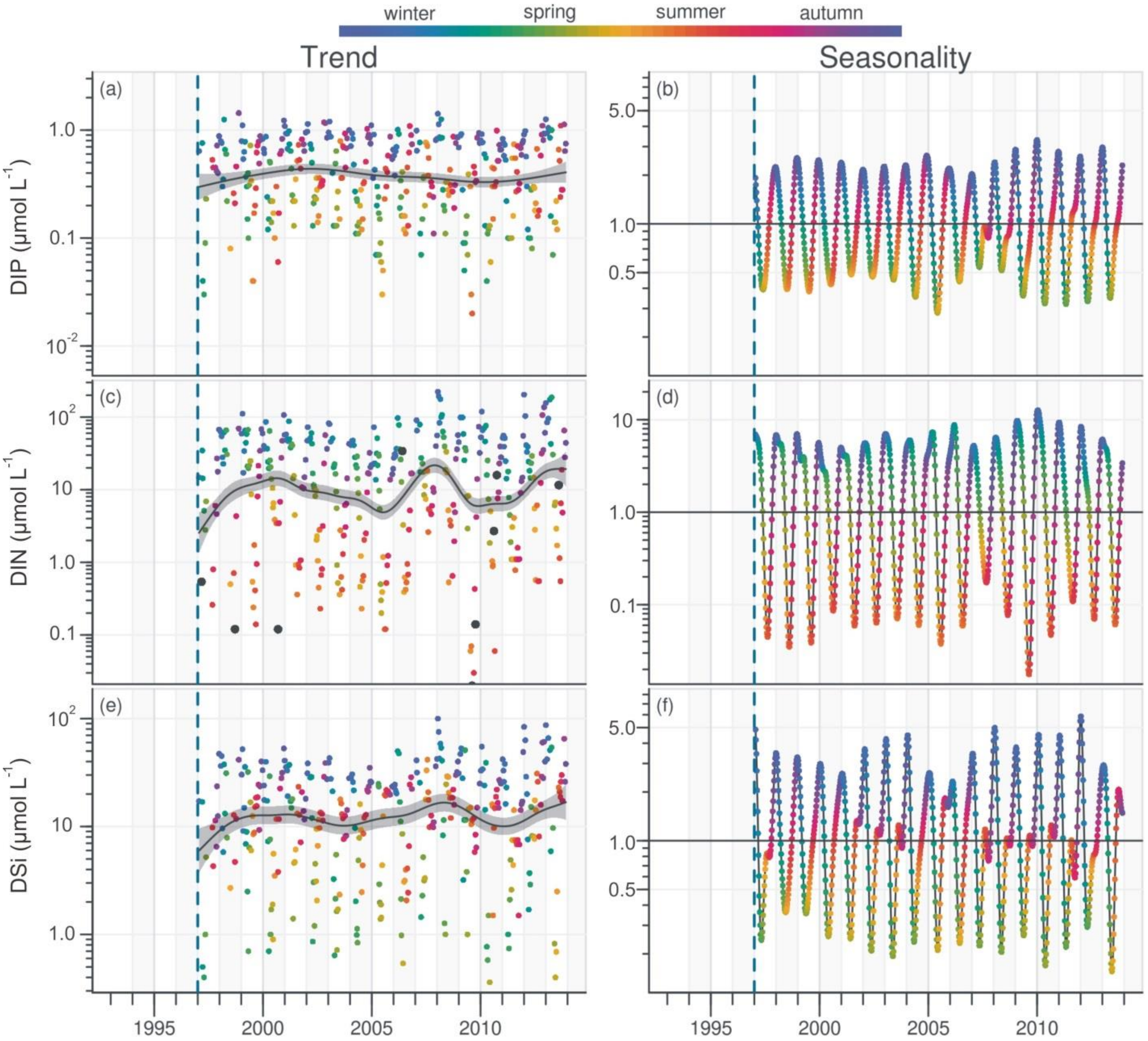


Figure 5: Long-term trend~~s~~ and seasonality of Chl *a* in the Loire (a, b). See Fig. 2 for details


winter
spring
summer
autumn
Trend
Seasonality
(a)
(b)
(c)
(d)
(e)
(f)
DIP (µmol L-1)
DIN (µmol L-1)
DSi (µmol L-1)
1.0
0.1
10-2
5.0
0.5
102
10
0.1
1995
2000
2005
2010

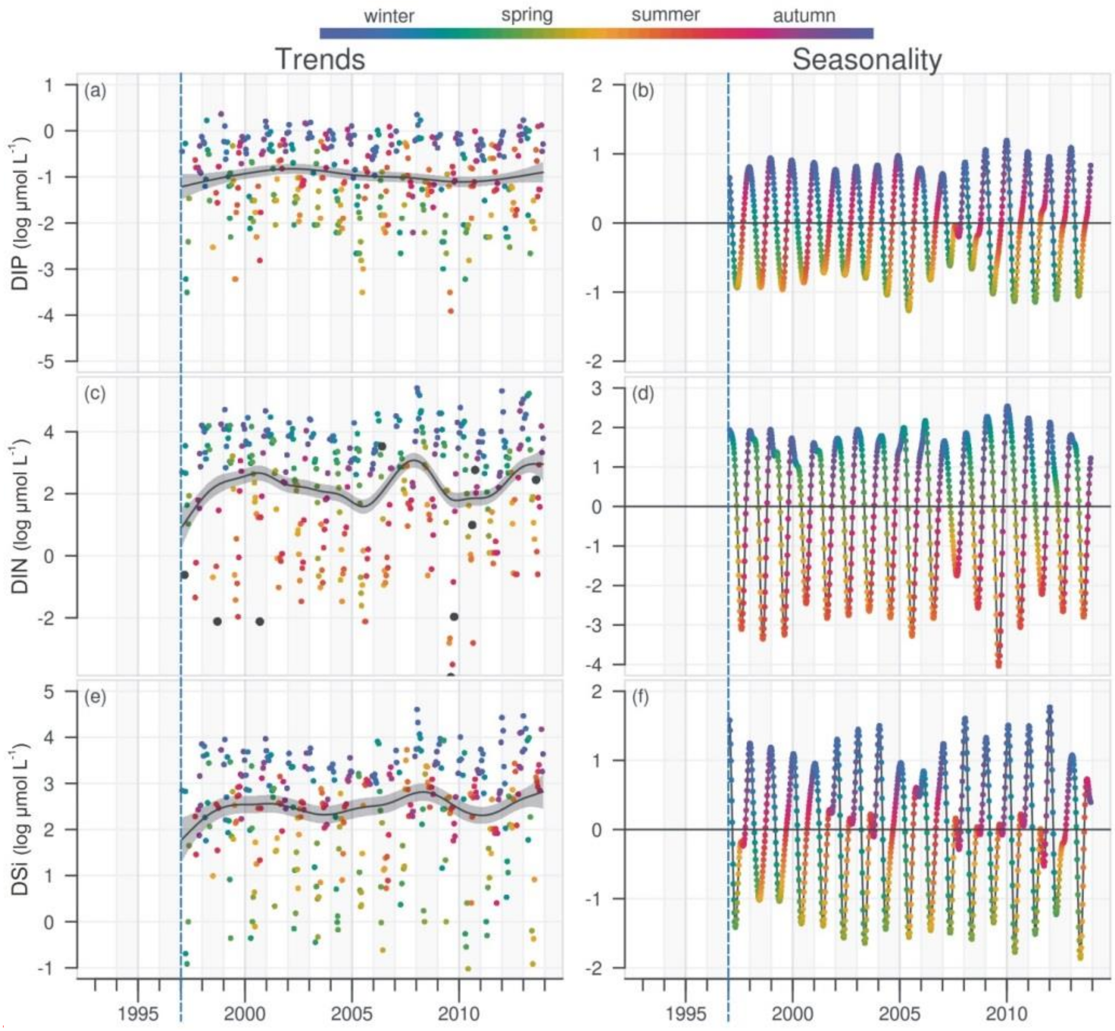
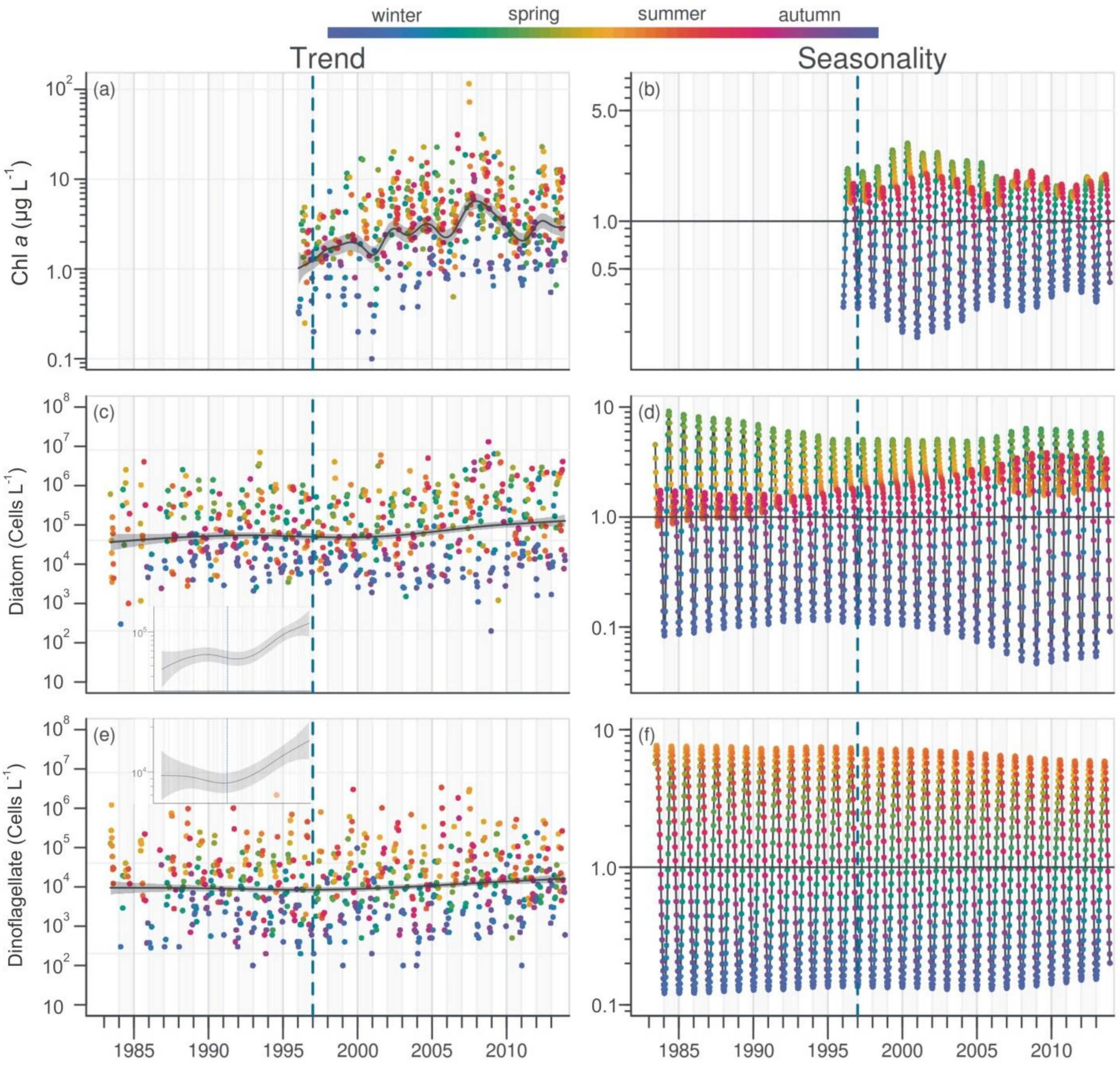


Figure 6: Long-term trend~~s~~ and seasonality of DIP (a, b), DIN (c, d) and DSi (e, f) in the V~~ilaine ~~B~~ay~~. Black dots represent data considered as outliers (see Section 2.4.). See Fig. 2 for details


winter
spring
summer
autumn
Trend
Seasonality
(a)
(b)
(c)
(d)
(e)
(f)
Chl *a* (µg L^{-1})
Diatom (Cells L^{-1})
Dinoflagellate (Cells L^{-1})
10^2
10
1.0
0.1
5.0
1.0
0.5
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10
0.1
10^5
10^4
1985
1990
1995
2000
2005
2010

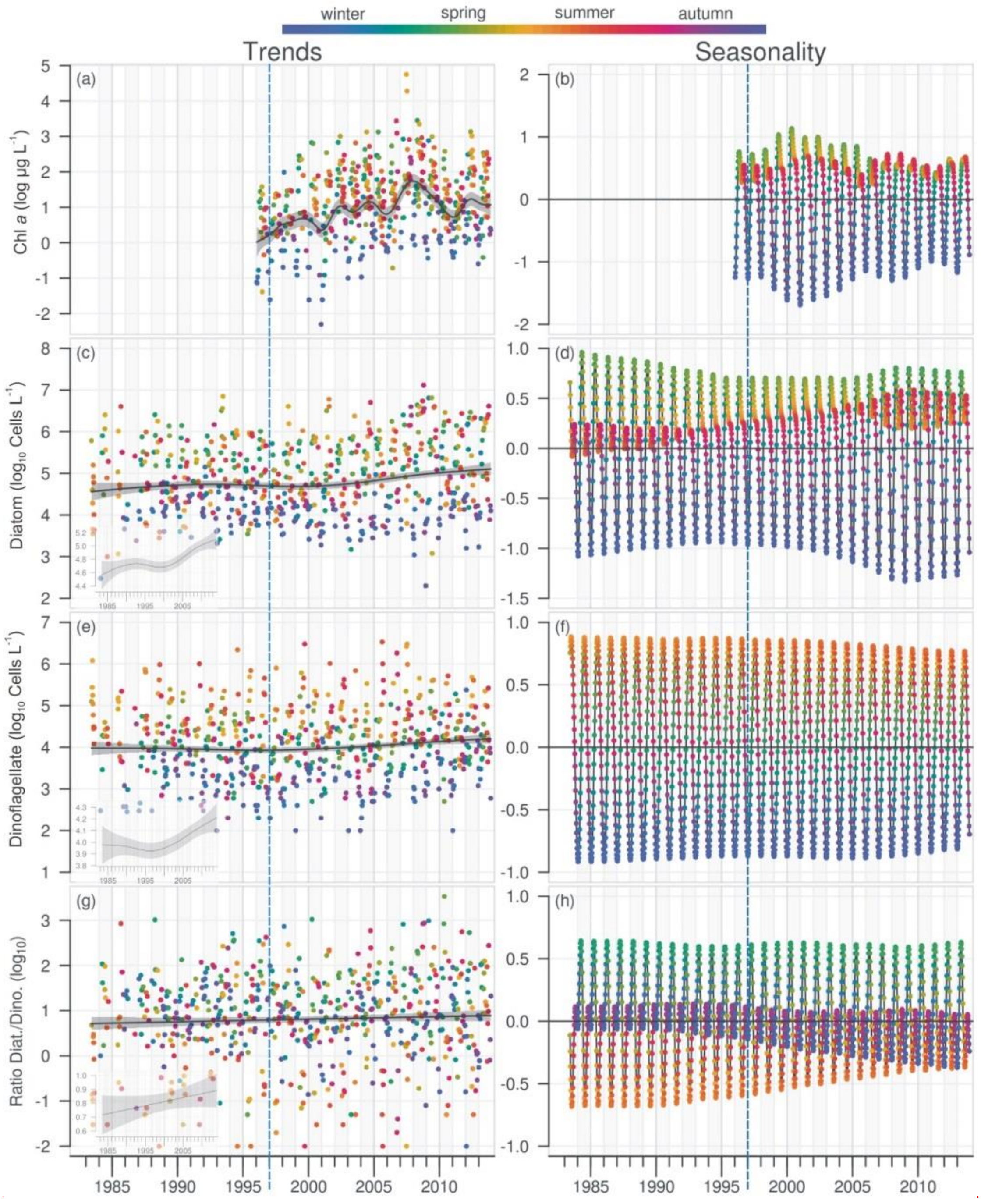

winter
spring
summer
autumn
Trends
Seasonality
(a)
(b)
(c)
(d)
(e)
(f)
(g)
(h)
Chl a (log µg L^{-1})
Diatom (log$_{10}$ Cells L^{-1})
Dinoflagellate (log$_{10}$ Cells L^{-1})
Ratio Diat./Dino. (log$_{10}$)
1985
1990
1995
2000
2005
2010

Figure 7: Long-term trend~~s~~ and seasonality of Chl *a* (a, b), diatom (c, d~~),~~) and dinoflagellate (e, f)~~ and diatom:dinoflagellate ratios~~ ~~(g, h)~~ in the VB. Insets ~~show~~ display trends of diatom and dinoflagellate abundances with optimal scale. See Fig. 2 for details

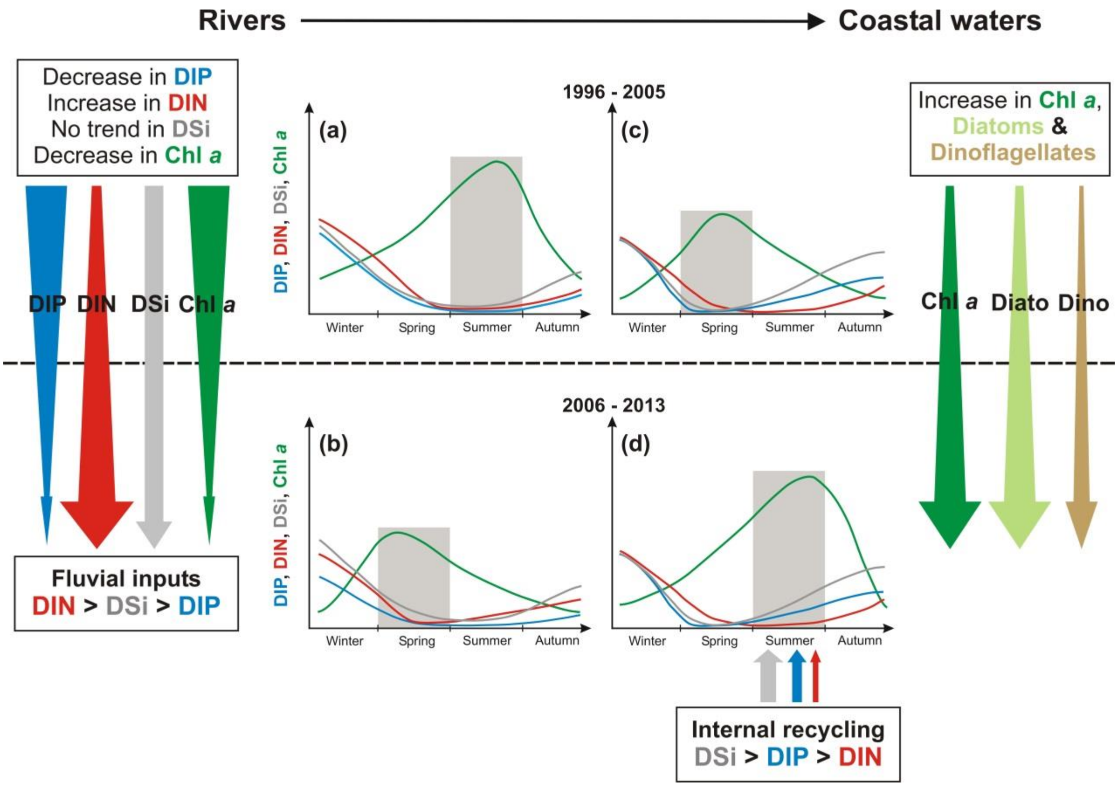


Figure 8: Graphical representation of the major changes in nutrient concentrations and phytoplankton in river (a, b) and the VB coastal waters~~ of VB~~ (c, d) for the period 1996-2005 (top) and 2006-2013 (bottom). Downward arrows and curves, representing respectively long-term trends and seasonal courses of eutrophication parameters in rivers and in the VB, were fitted according to results. Shaded areas underline the season of maximum Chl *a*. Internal benthic nutrient inputs (upward arrows) were fitted according to the measurement of benthic fluxes in summer 2015 (~~see Table 4~~Ratmaya, 2018)

Table 1: Statistical results from Mann-Kendall test performed DLM trend components of eutrophication parameters in rivers and in the VB coastal waters ~~on time series of eutrophication parameters in rivers~~ for the common period 1997-2013. If the test was significant at $p<0.05$, differences of the Sen's robust line between the beginning and the end of the period (17 years) were calculated. ~~Trends were calculated as a difference between values in the beginning and in the end of the period (16 years) of the Kendall-Theil robust line, only if the test was significant at $p<0.05$ (in bold).~~ Values in parentheses are percentages of ~~trends~~ changes relative to the initial values of the ~~Kendall-Theil~~Sen's robust line. Increasing or decreasing trends are indicated by + and – signs respectively. Cells were left blank when tests were not applicable. NS = non-significant~~Symbols indicate increasing or decreasing trends~~

Site	Discharge (m^3 s^{-1})		DIP (µmol L^{-1})		DIP loads (mol s^{-1})		DIN (µmol L^{-1})		DIN loads (mol s^{-1})		DSi (µmol L^{-1})		Chl *a* (µg L^{-1})		Diatoms (Cells L^{-1})		Dinoflagellates (Cells L^{-1})	
	p	Change (%)	*p*	Change (%)	*p*	Change (%)	*p*	Change (%)	*p*	Change (%)	*p*	Change (%)	*p*	Change (%)	*p*	Change (%)	*p*	Change (%)
Loire	0.01	− 94 (16%)	<0.001	− 0.85 (47%)	<0.001	− 0.60 (52%)	0.63	NS	0.42	NS			<0.001	− 54 (93%)				
Vilaine	0.02	− 8.7 (23%)	<0.001	− 1.9 (75%)	<0.001	− 0.09 (88%)	<0.001	− 71 (21%)	<0.001	− 4.6 (38%)			<0.001	− 12 (76%)				
VB			<0.001	− 0.05 (13%)			0.01	+ 3.2 (40%)			<0.001	+ 3.6 (34%)	<0.001	+ 2.1 (126%)	<0.001	+ 90*10^3 (227%)	<0.001	+ 8*10^3 (108%)

Table 2: Statistical results of modified Mann-Kendal test performed on DLM seasonal components of eutrophication parameters in rivers and in the VB for the common period 1997–2013. If the test was significant at *p*<0.05, percentages of changes relative to the initial values of the Sen's robust line were calculated. Increasing or decreasing trends are indicated by + and – signs respectively. Cells were left blank when tests were not applicable. NS = non-significant~~Summary of statistical results from Mann-Kendall test performed on time series of eutrophication parameters in VB for the period 1997-2013. See Table 1 for details~~

Site/ Season	Discharge (m^3 s^{-1})		DIP (µmol L^{-1})		DIP loads (mol s^{-1})		DIN (µmol L^{-1})		DIN loads (mol s^{-1})		DSi (µmol L^{-1}		Chl *a* (µg L^{-1})		Diatoms (Cells L^{-1})		Dinoflagellates (Cells L^{-1})	
	p	%	*p*	%	*p*	%	*p*	%	*p*	%	*p*	%	*p*	%	*p*	%	*p*	%
Loire																		
Winter	0.63	NS	0.04	− 23%	<0.01	− 41%	0.02	− 24%	<0.01	− 40%			<0.001	+ 190%				
Spring	0.50	NS	<0.001	− 28%	0.02	− 33%	0.21	NS	0.49	NS			<0.001	+ 283%				
Summer	0.60	NS	<0.001	+ 33%	<0.001	+ 59%	<0.01	+ 55%	0.01	+ 69%			0.09	NS				
Autumn	0.98	NS	<0.01	+ 35%	0.26	NS	0.29	NS	0.92	NS			<0.001	− 82%				
Vilaine																		
Winter	0.23	NS	0.02	− 17%	0.07	NS	0.90	NS	0.11	NS			<0.01	+ 97%				
Spring	0.93	NS	0.06	NS	0.07	NS	0.99	NS	0.56	NS			<0.001	+ 63%				
Summer	0.26	NS	<0.001	+ 9.4%	0.09	NS	0.29	NS	0.28	NS			<0.001	− 41%				
Autumn	0.97	NS	0.51	NS	0.40	NS	0.66	NS	0.69	NS			0.01	− 44%				
VB																		
Winter			0.73	NS			0.03	+ 32%			0.329	NS	0.11	NS	0.85	NS	0.05	NS
Spring			<0.001	− 30%			0.10	NS			0.086	NS	<0.001	− 36%	0.93	NS	0.83	NS
Summer			<0.001	+ 80%			0.17	NS			0.085	NS	0.19	NS	<0.001	+ 43%	<0.001	− 23%
Autumn			0.94	NS			0.76	NS			0.647	NS	0.37	NS	0.27	NS	0.87	NS

Table 3: Spearman's rank correlations between annual median values of river discharge, nutrient concentrations and phytoplankton biomass in the Loire~~ and ~~, Vilaine~~ Rivers~~ and ~~in ~~the VB ~~between 1980 and 2013~~for the common period (*n* = 17). Asterisks designate significant correlations (*p<0.001, **p<0.01, *p<0.05)~~. Relatively strong correlations (-0.50 ≥ r ≥ +0.50) are in bold and the number of data points are in brackets~~**

	Loire discharge	Vilaine discharge	DIN Loire	DIP Loire	Chl *a* Loire	DIN Vilaine	DIP Vilaine	Chl *a* Vilaine	DIN VB	DIP VB	DSi VB	Chl *a* VB
Loire discharge	1.00											
Vilaine discharge	0.88***	1.00										
DIN Loire	0.52*	0.39	1.00									
DIP Loire	0.51*	0.43	0.44	1.00								
Chl *a* Loire	-0.08	-0.06	0.25	0.35	1.00							
DIN Vilaine	0.33	0.47	0.02	0.55*	0.59*	1.00						
DIP Vilaine	0.16	0.24	0.23	0.77**	0.65*	0.54	1.00					
Chl *a* Vilaine	-0.21	-0.28	0.31	0.20	0.64**	0.04	0.35	1.00				
DIN VB	0.78**	0.74**	0.36	0.35	-0.10	0.29	-0.01	-0.20	1.00			
DIP VB	0.13	-0.09	0.07	0.38	0.05	0.11	0.29	0.19	-0.12	1.00		
DSi VB	0.55*	0.41	0.35	0.08	-0.48	-0.17	-0.51	-0.31	0.63*	-0.02	1.00	
Chl *a* VB	0.11	0.17	-0.14	-0.48	-0.61*	-0.34	-0.58*	-0.50*	0.25	-0.45	0.33	1.00

	~~Loire discharge~~	~~Vilaine discharge~~	~~DIN Loire~~	~~DIP Loire~~	~~Chl *a* Loire~~	~~DIN Vilaine~~	~~DIP Vilaine~~	~~Chl *a* Vilaine~~	~~DIN VB~~	~~DIP VB~~	~~DSi VB~~	~~Chl *a* VB~~
~~Loire discharge~~	~~1.00~~											
~~Vilaine discharge~~	~~**0.81*** (34)**~~	~~1.00~~										
~~DIN Loire~~	~~0.40* (34)~~	~~0.46** (34)~~	~~1.00~~									
~~DIP Loire~~	~~0.10 (24)~~	~~0.03 (24)~~	~~-0.06 (24)~~	~~1.00~~								
~~Chl *a* Loire~~	~~-0.07 (34)~~	~~-0.03 (34)~~	~~-0.10 (34)~~	~~**0.60** (24)**~~	~~1.00~~							
~~DIN Vilaine~~	~~-0.09 (34)~~	~~0.15 (34)~~	~~0.20 (34)~~	~~**0.67** (24)**~~	~~0.33 (34)~~	~~1.00~~						
~~DIP Vilaine~~	~~-0.21 (31)~~	~~-0.14 (31)~~	~~-0.44* (31)~~	~~**0.87*** (21)**~~	~~**0.69*** (31)**~~	~~0.24 (31)~~	~~1.00~~					
~~Chl *a* Vilaine~~	~~-0.05 (34)~~	~~-0.15 (34)~~	~~-0.12 (34)~~	~~0.41* (24)~~	~~**0.63*** (34)**~~	~~-0.07 (34)~~	~~0.42* (31)~~	~~1.00~~				
~~DIN VB~~	~~**0.78** (17)**~~	~~**0.74** (17)**~~	~~0.36 (17)~~	~~0.35 (17)~~	~~-0.10 (17)~~	~~0.29 (17)~~	~~-0.01 (14)~~	~~-0.20 (17)~~	~~1.00~~			
~~DIP VB~~	~~0.13 (17)~~	~~-0.09 (17)~~	~~0.07 (17)~~	~~0.38 (17)~~	~~0.05 (17)~~	~~0.11 (17)~~	~~0.29 (14)~~	~~0.19 (17)~~	~~-0.12 (17)~~	~~1.00~~		
~~DSi VB~~	~~**0.55* (17)**~~	~~0.41 (17)~~	~~0.35 (17)~~	~~0.08 (17)~~	~~-0.48 (17)~~	~~-0.17 (17)~~	~~-0.51 (14)~~	~~-0.31 (17)~~	~~**0.63* (17)**~~	~~-0.02 (17)~~	~~1.00~~	
~~Chl *a* VB~~	~~0.20 (18)~~	~~0.25 (18)~~	~~0.04 (18)~~	~~**-0.56* (18)**~~	~~**-0.67** (18)**~~	~~-0.32 (18)~~	~~**-0.66* (15)**~~	~~**-0.58* (18)**~~	~~0.25 (17)~~	~~-0.45 (17)~~	~~0.33 (17)~~	~~1.00~~

~~**Table 4: Comparison of nutrient inputs from sediments (benthic fluxes) with inputs from the Loire and the Vilaine Rivers in VB during summer 2015 (tons)**~~

~~Nutrients~~	~~Benthic inputs~~ [a]	~~Vilaine inputs~~ [b]	~~Loire inputs~~ [b]
~~DIN-N~~	~~1078~~	~~1286~~	~~1183~~
~~DIP-P~~	~~211~~	4	~~15~~
~~DSi-Si~~	~~8868~~	~~707~~	~~1853~~

[a] **~~Benthic fluxes were measured in June, August and September 2015 in VB at MOLIT station (Retho et al. 2016) using sediment core incubation method (Ratmaya et al., in prep) and extrapolated to the area of VB (220 km^2; Ehrhold 2014).~~**

[b] **~~Fluvial nutrient inputs were calculated from monthly nutrient concentrations and monthly averaged river discharges (see Material and Methods). Inputs from the Loire were divided by 10 to account for the dilution effect due to the distance (Ménesguen and Dussauze 2015). Note that inputs from the Vilaine calculated from the measurement station upstream, did not take into account the closing of the dam (Traini et al. 2015).~~**