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 (i.e., phytoplankton biomass) during recent decades was examined in the coastal
15 waters of 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.
The reduction in dissolved riverine inorganic phosphorus concentrations (DIP) from the early 1990s led to the decrease in
their Chl *a* levels. However, dissolved inorganic nitrogen concentrations (DIN) decreased only slightly in the Vilaine and
20 actually increased in the Loire, especially during summer. Simultaneously, phytoplankton in the VB has undergone profound
changes with: 1) increase in biomass, 2) change in the position of the annual peak from spring to summer, and 3) increase in
diatom:dinoflagellate ratios, especially in summer. The increase in phytoplankton biomass in VB, particularly in summer,
was probably due to increased DIN loads from the Loire, sustained by internal regeneration of DIP and dissolved silicate
from sediments. This long-term ecosystem-scale analysis reports the consequence of nutrient management scenarios focused
25 solely on P reduction. Freshwater ecosystems upstream reveal successful recoveries through the control of P alone, while
eutrophication continues to increase downstream, especially during the period of N limitation. Therefore, nutrient
management strategies, paying particular attention to diffuse N-sources, are required to control eutrophication in receiving
coastal waters.

Keywords: eutrophication, nutrients, phytoplankton, 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 than for N (Grizzetti et al., 2012) but still led to reduction in phytoplankton biomass (Istvánovics and Honti, 2012; Romero et al., 2013). This result is consistent with the idea that reducing P alone, and not N, can mitigate eutrophication of freshwater ecosystems (Schindler et al., 2008). 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 (Williams et al., 2010), the Seine Bay (Romero et al., 2013) and many other coastal ecosystems (see Cloern, 2001).

The Loire River, alongside the Vilaine River, is part of these major European rivers whose phytoplankton biomass and P concentrations have decreased since the early 1990s, but without a diminution in N concentrations (Romero et al., 2013; Minaudo et al., 2015). 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, Vilaine Bay (VB; FRGC44) is one of the European Atlantic coastal ecosystems most sensitive to eutrophication (Chapelle et al., 1994; Ménesguen et al., 2014). 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 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 VB, is usually limited by P in spring and by N in summer.

The present study examined the long-term changes (trend and seasonality) of phytoplankton biomass and nutrient concentrations in the coastal waters of VB, 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 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 agricultural, producing

5 two-thirds of the national livestock and half the cereal production (Aquilina et al., 2012; Larroudé et al., 2013). 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 water residence time in the

10 bay 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).

2.2 Long-term monitoring dataset: Rivers, Vilaine Bay, Bay of Biscay

15 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

20 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 in Table S1. Briefly, nutrient

25 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 carried out essentially for organisms whose size is >20 μm (i.e., micro-phytoplankton), also 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).

2.3 Data pre-processing

Most of DIN concentrations measured in fresh and marine waters consisted mainly of nitrate (>90%; Garnier et al., 2010;
5   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
10   (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
15   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 Time-series analyses

The time-series were modeled using Dynamic Linear Models (DLM; West and Harrison, 1997) with the *dlm* package (Petris,
20   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 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
25   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 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

5   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).

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

10   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.

In order to corroborate the overall trends observed by DLM, a modified non-parametric Mann-Kendall (MK) test (Hamed

15   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).

20   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 River trends

The daily discharge of the Loire varied between 157 and 3,425 m^3 s^{-1} from 1980 to 2013 with an overall median of

25   614 m^3 s^{-1}. The DLM trend plot of the Loire discharge displayed oscillations with periodicities of 6-7 years and no noticeable overall trend (Fig. 2a). The MK test performed for the period of 1997-2013 revealed a decrease of 94 m^3 s^{-1} over 16 years (Table 1). No marked change was observed in the seasonality (Fig. 2b), as confirmed by MK test with no significant trend whatever the season (Table 1). The Vilaine discharge (median of 36 m^3 s^{-1} for the period of 1980-2013) represented only 6 %

of the Loire discharge and displayed similar trend and seasonality to those of the Loire (Fig. S2), as highlighted by the significant correlation between their annual medians (Table 3).

Annual median DIP in the Loire decreased from around 2.5 µmol L^{-1} in the early 1990s to values near 1.5 µmol L^{-1} in 2010s (Fig. 3a), which was confirmed by MK test for the period of 1997-2013 (Table 1). The seasonality plot indicated a shift in
5 position of annual DIP minimum from summer to spring, observed particularly between 1997 and 2013 (Fig. 3b). The MK test confirmed this seasonality pattern with a significant negative trend for spring values and a significant positive trend for summer values (Table 1). The trend of DIP loads from the Loire displayed oscillations reflecting the influence of river discharge (Fig 3c), with a significant decreasing trend in their annual medians (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
10 summer and winter (Fig. 3d). Patterns of DIP and DIP loads for the Vilaine were similar to those for the Loire (Fig. S3, Table 1), as indicated by a significant correlation between annual medians of DIP in the two rivers (Table 3).

DIN in the Loire displayed 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}. The MK test for DIN in the Loire did not indicate a significant trend from
15 1997 to 2013 (Table 1). However, the seasonality plot of DIN in the Loire was marked by a decrease in the seasonal amplitude from the early 1990s (Fig. 4b), with a significant positive trend for summer values and a significant negative trend for winter values (Table 1). The DIN loads from the Loire displayed similar trend and seasonality to those of DIN (Figs. 4c, 4d, Table 1). The DLM trend of DIN in the Vilaine revealed an increase before the mid-1990s and a slight decrease subsequently (Fig. S4a). The latter was significant according to the MK test (Table 1). The seasonality plot of DIN
20 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, S4d, Table 1).

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).

25 The Chl a trend in the Loire was stable between 1980 and 2000 with annual medians around 60 µg L^{-1} and then decreased strongly from 2000 to reach median values below 10 µg L^{-1} from 2010 onwards (Fig. 5a). A significant decrease was also indicated by MK test for the period of 1997-2013 (Table 1). The decreasing trend in annual Chl a in the Loire was accompanied by a shift in position of the annual maximum from late summer before 2000 to spring afterwards (Fig. 5b). This seasonality pattern was confirmed by MK test with a significant negative trend for autumn values and significant
30 positive trend for winter and spring values (Table 1). DLM and MK results for Chl a in the Vilaine revealed similar trend

and seasonality to those in the Loire (Fig. S6, Table 1). Annual medians of Chl *a* and DIP in the two rivers correlated significantly with each other (Table 3).

3.2 Vilaine Bay (VB) trends

For overall studied period, annual DIP medians in VB were close to 0.5 µmol L^{-1}. The DLM trend seemed to be stable

5   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 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

10   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). 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},

15   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).

Annual Chl *a* medians in VB were negatively correlated with DIP and Chl *a* from both rivers (Table 3). Chl *a* in VB

20   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).

The DLM trend of diatom abundances in VB suggested an increasing trend between 1983 and 2013 (Fig. 7c), as confirmed

25   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

5 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

4.1 The Loire/Vilaine – Vilaine Bay continuum

10 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

15 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.

20 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

25 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 Phytoplankton in river and coastal waters

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).

4.3 Nutrient trends in rivers

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).

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}

5   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

10   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

15   pointed out by Grizzetti et al. (2012) for most large rivers on Atlantic coasts. (Brzezinski, 1985)

4.4 Nutrient trends in coastal waters

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

20   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

25   phytoplankton in VB must be interpreted essentially according to variations in riverine nutrient inputs, especially those from the Loire (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

30   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.

5   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

10 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

15 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 Implication for nutrient management

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

20 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

25 (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

30 et al., 2009; Paerl, 2009).

5. Conclusion and perspective

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

5 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

10 shellfish area (Désaunay et al., 2007; 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

15 Phytoplankton and Hydrology in coastal waters (http://www.seanoe.org/data/00361/47248/), 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

20 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

25 The authors declare that they have no conflict of interest.

Acknowledgements

This study was funded by The Loire – Brittany Water Agency (AELB). Authors are grateful to Sylvain Jolly from AELB for providing datasets of the Vilaine and the Loire. We thank IFREMER-LER/MPL staff for their technical contribution, especially Karine Collin, Yoann Le Merrer, Mireille Fortune, Michël Retho, Raoul Gabellec, Jacky Chauvin, Isabelle

5 Truquet and Anne Schmitt. We thank Alice Mellor for the English corrections. Authors are grateful to Drs. F. Gerald Plumley and Anniet M. Laverman for kindly reviewing before submission. Authors acknowledge IFREMER and the Regional Council of the Région des Pays de la Loire for the PhD funding of W. Ratmaya.

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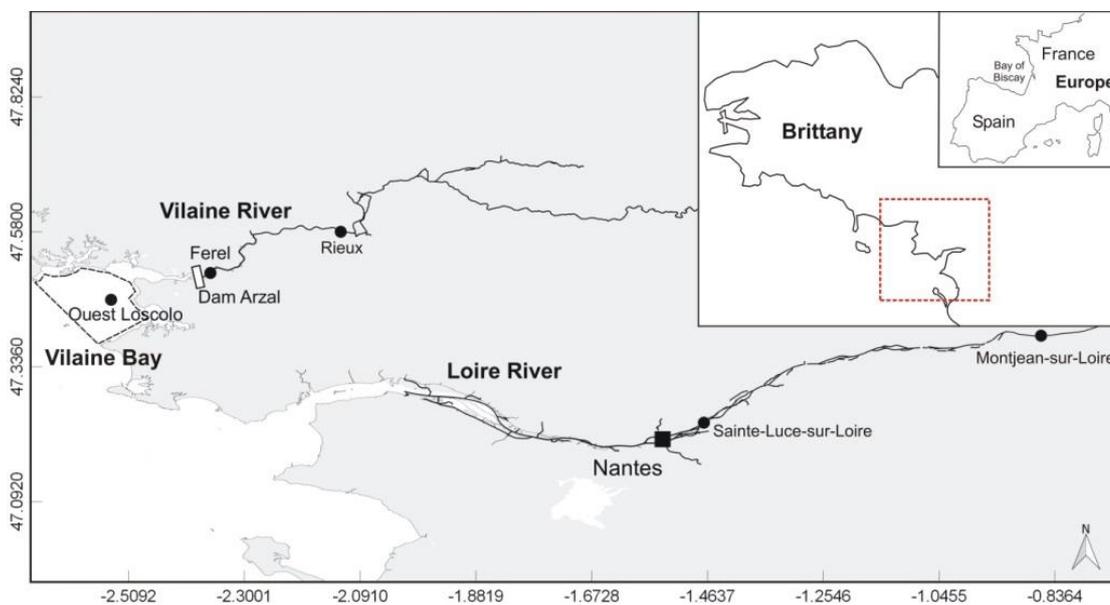
Figure 1: Map of the area studied showing Loire and Vilaine rivers and delimitation of Vilaine Bay (inset red dotted line). Black dots mark the sampling and gauging stations cited
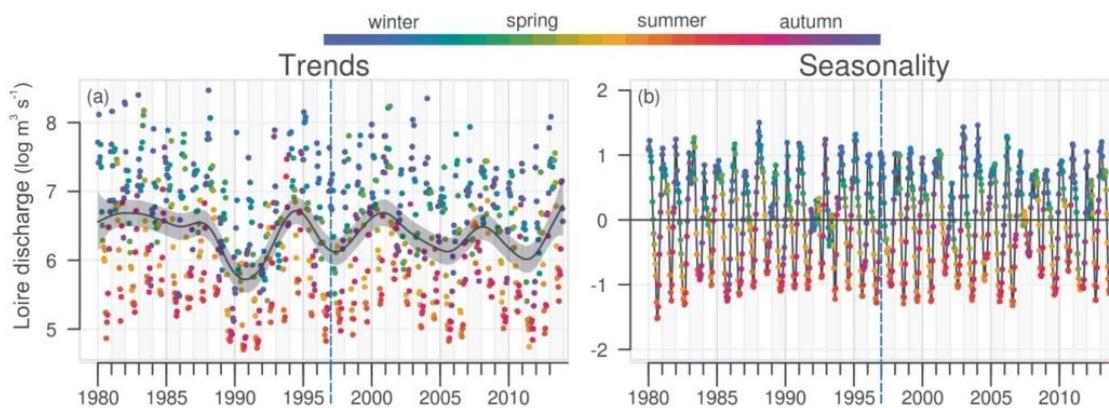
Figure 2: Long-term trends and seasonality of the Loire discharges (a, b). Dark grey lines represent DLM trends. Shaded areas indicate the 95 % confidence interval. Each dot in the trend plot (left) represents an observed value, those in the seasonality plot (right) represent values estimated by the model. Dashed vertical blue line corresponds to the period for MK test (1997-2013)
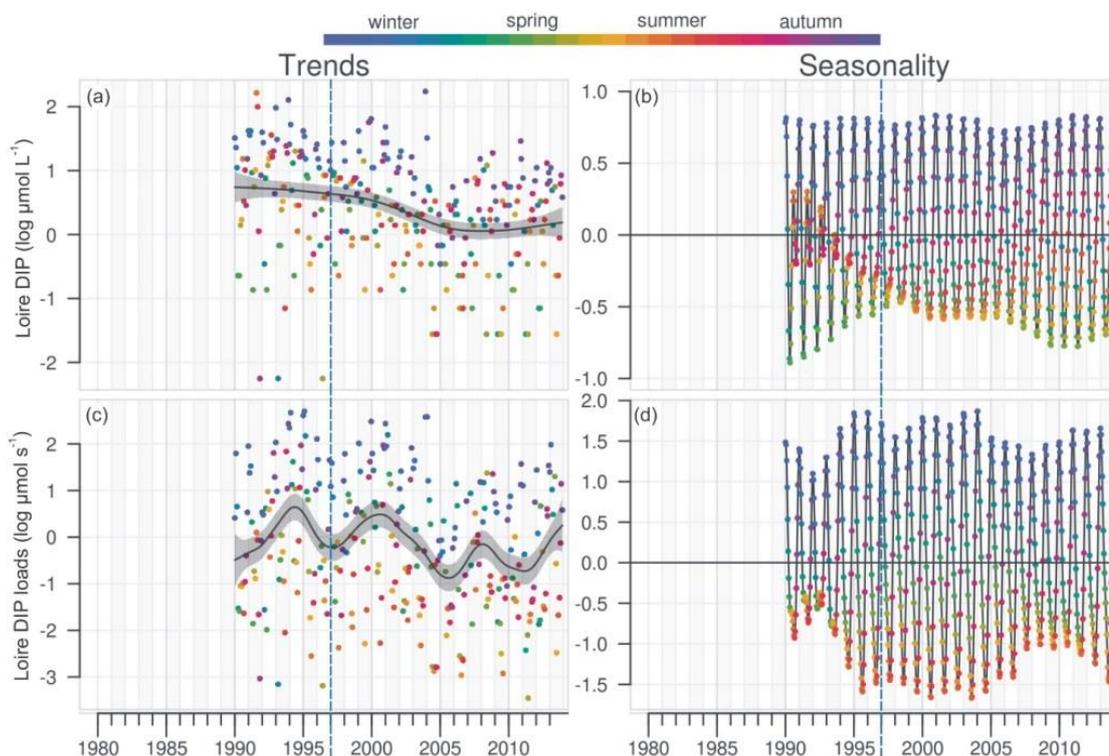
Figure 3: Long-term trends and seasonality of DIP in the Loire (a, b) and DIP loads from the Loire (c, d). Note: analytical problems of DIP measurements before 1990 (see Section 2.4.). See Fig. 2 for details
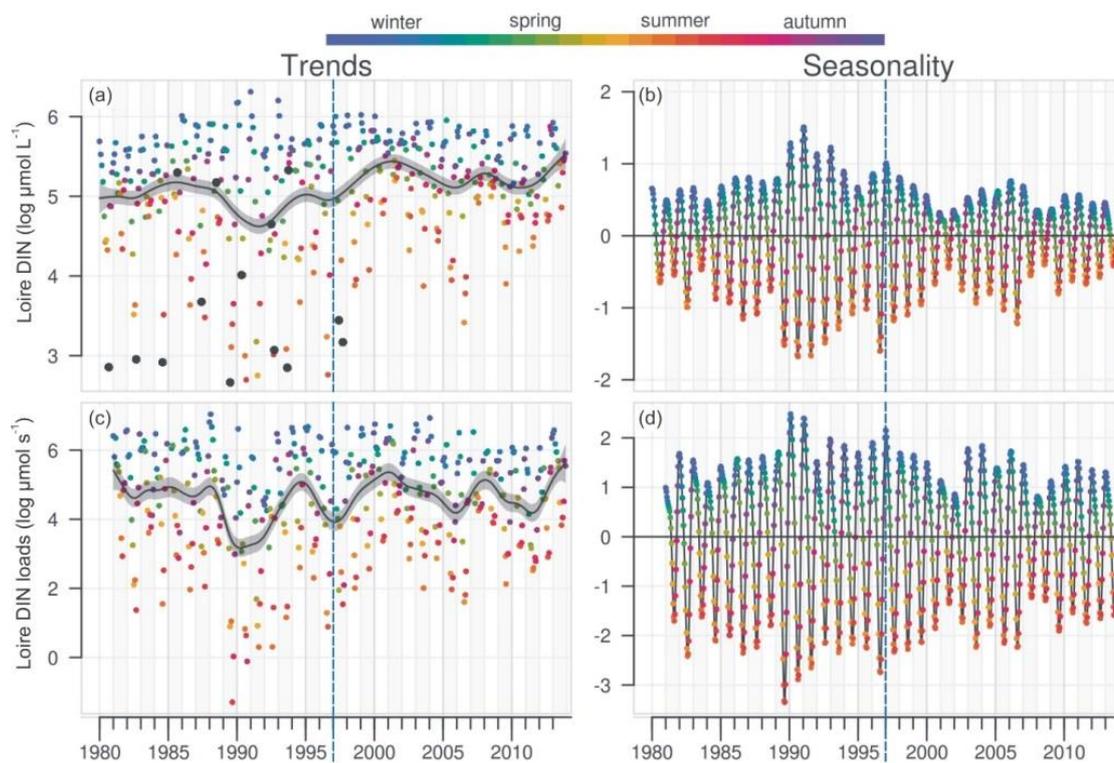
Figure 4: Long-term trends and seasonality of DIN in the Loire (a, b) and DIN loads from the Loire (c, d). Black dots represent data considered as outliers (see Section 2.4.). See Fig. 2 for details
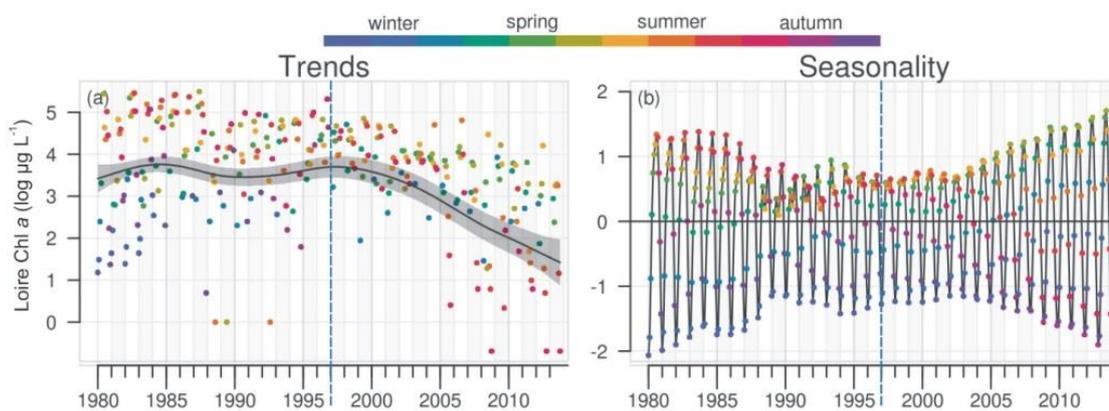
Figure 5: Long-term trends and seasonality of Chl *a* in the Loire (a, b). See Fig. 2 for details
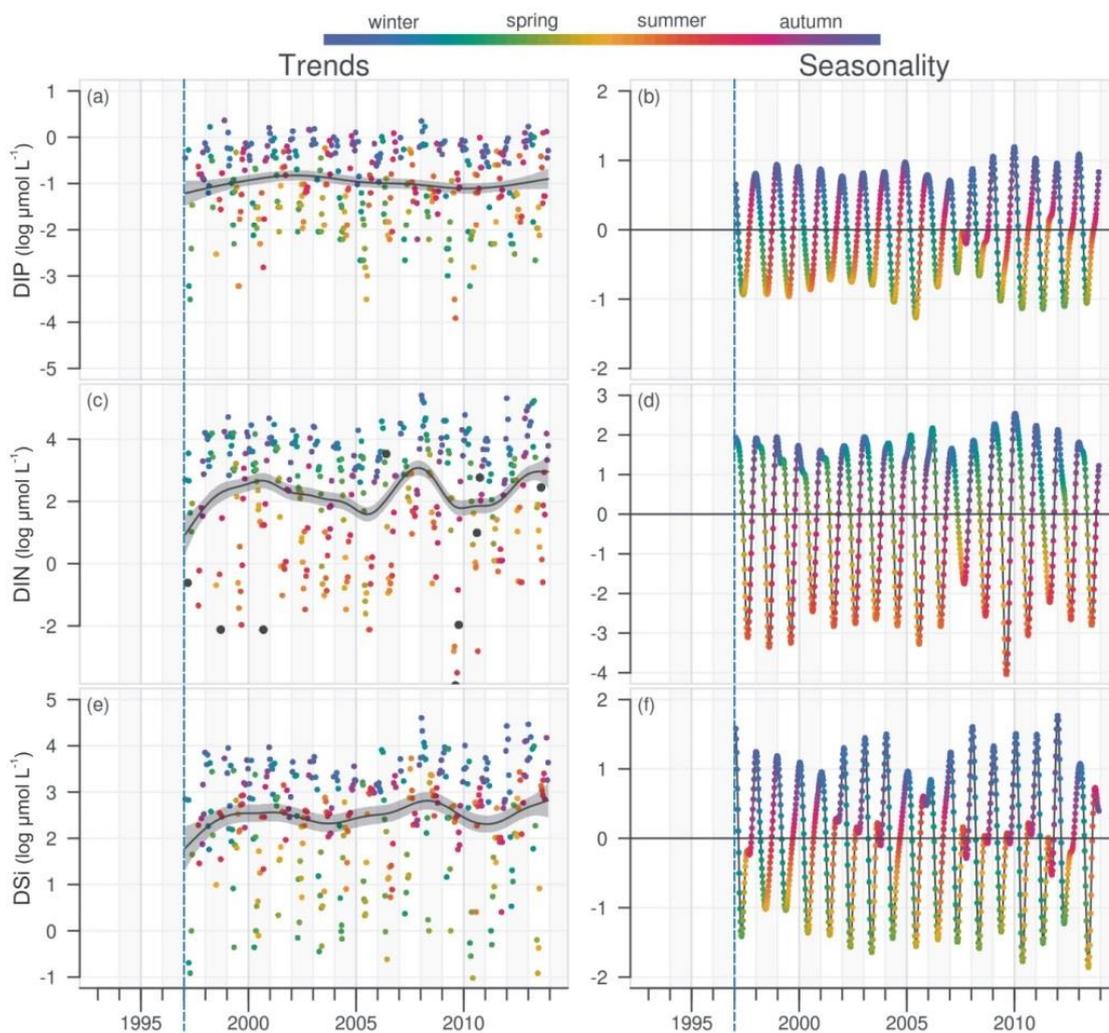
Figure 6: Long-term trends and seasonality of DIP (a, b), DIN (c, d) and DSi (e, f) in Vilaine Bay. Black dots represent data considered as outliers (see Section 2.4.). See Fig. 2 for details
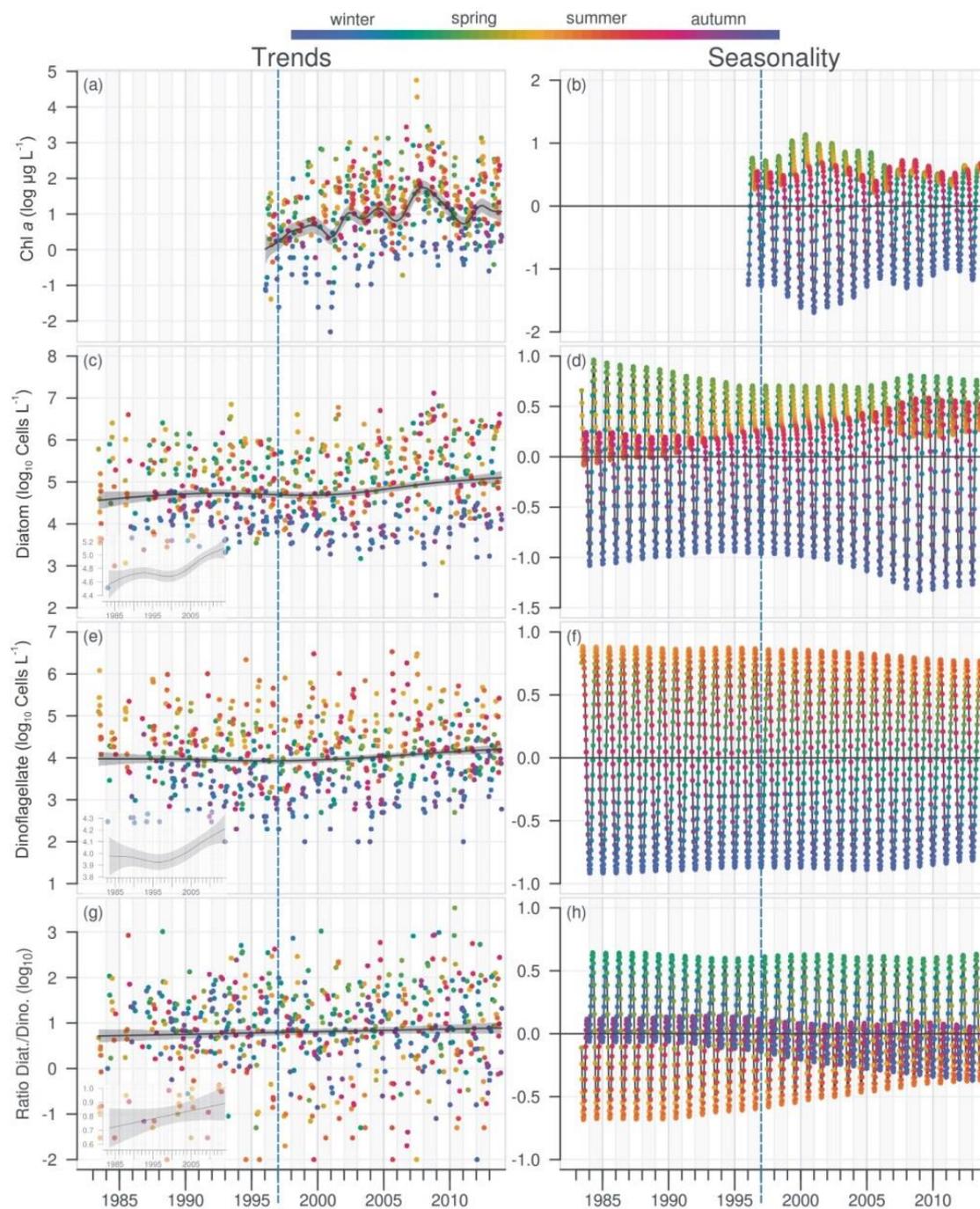
Figure 7: Long-term trends and seasonality of Chl *a* (a, b), diatom (c, d), dinoflagellate (e, f) and diatom:dinoflagellate ratios (g, h) in VB. Insets show trends with optimal scale. See Fig. 2 for details
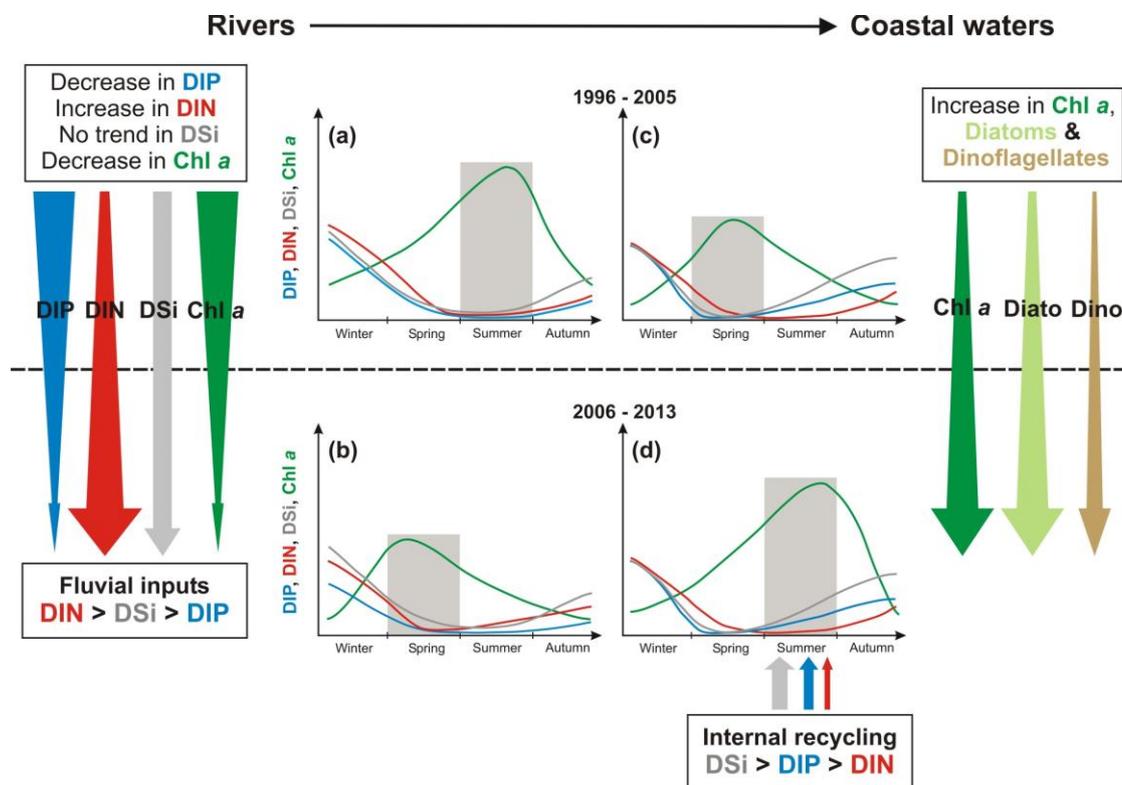
Figure 8: Graphical representation of the major changes in nutrient concentrations and phytoplankton in river (a, b) and coastal waters of VB (c, d) for the period 1996-2005 (top) and 2006-2013 (bottom). Downward arrows and curves, representing respectively long-term trends and seasonal courses of eutrophication parameters in rivers and in VB, were adjusted according to results. Shaded areas underline the season of maximum Chl *a*. Internal benthic nutrient inputs (upward arrows) were adjusted according to the measurement of benthic fluxes in summer 2015 (see Table 4)

Table 1: Summary of statistical results from Mann-Kendall test performed on time series of eutrophication parameters in rivers for the period 1997-2013. 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 relative to the initial values of the Kendall-Theil robust line. Symbols indicate increasing or decreasing trends

Site/Season	River discharge		DIP		Flux DIP		DIN		Flux DIN		Chl a	
	p value	Trend ($m^3 s^{-1}$)	p value	Trend ($\mu mol\,L^{-1}$)	p value	Trend ($\mu mol\,s^{-1}$)	p value	Trend ($\mu mol\,L^{-1}$)	p value	Trend ($\mu mol\,s^{-1}$)	p value	Trend ($\mu g\,L^{-1}$)
Loire River												
Year	**0.014**	− 94 (16%)	**<0.001**	− 0.85 (47%)	**<0.001**	− 0.60 (52%)	0.630	NS	0.422	NS	**<0.001**	− 54 (93%)
Winter	0.626	NS	**0.037**	−	**0.007**	−	**0.025**	−	**0.005**	−	**<0.001**	+
Spring	0.499	NS	**<0.001**	−	**0.020**	−	0.209	NS	0.490	NS	**<0.001**	+
Summer	0.603	NS	**<0.001**	+	**<0.001**	+	**0.002**	+	**0.014**	+	0.095	NS
Autumn	0.986	NS	**0.004**	+	0.264	NS	0.290	NS	0.922	NS	**<0.001**	−
Vilaine River												
Year	**0.021**	− 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%)
Winter	0.227	NS	**0.022**	−	0.074	NS	0.900	NS	0.114	NS	**0.008**	+
Spring	0.931	NS	0.059	NS	0.067	NS	0.996	NS	0.508	NS	**<0.001**	+
Summer	0.261	NS	**<0.001**	+	0.090	NS	0.293	NS	0.286	NS	**<0.001**	−
Autumn	0.969	NS	0.512	NS	0.400	NS	0.658	NS	0.694	NS	**0.015**	−

Table 2: 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	DIP		DIN		DSi		Chl *a*		Diatoms		Dinoflagellates		Diatoms/ Dinoflagellates	
	p value	Trend (µmol L^{-1})	*p* value	Trend (µmol L^{-1})	*p* value	Trend (µmol L^{-1})	*p* value	Trend (µg L^{-1})	*p* value	Trend (Cells L^{-1})	*p* value	Trend (Cells L^{-1})	*p* value	Trend
Vilaine Bay														
Year	**<0.001**	− 0.35 (13%)	**0.009**	+ 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%)	**<0.001**	+
Winter	0.729	NS	**0.034**	+	0.329	NS	0.112	NS	0.846	NS	0.054	NS	0.125	NS
Spring	**<0.001**	−	0.099	NS	0.086	NS	**<0.001**	−	0.931	NS	0.826	NS	0.966	NS
Summer	**<0.001**	+	0.166	NS	0.085	NS	0.187	NS	**<0.001**	+	**<0.001**	−	**<0.001**	+
Autumn	0.937	NS	0.764	NS	0.647	NS	0.367	NS	0.274	NS	0.875	NS	**0.022**	−

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 VB between 1980 and 2013. 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	**0.81*** (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).

5   [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).