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

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Table S1: Available datasets for all sites of this study

Sites/ Parameter		Loire	Vilaine	Method	References	Vilaine Bay	Method	References
DIN (μmol L^{-1})	Nitrate	1980-2013 (monthly)	1980-2013 (monthly)	Flow analysis - Photometry	NF-EN ISO 13395	1997-2013 (bimonthly)	Flow analysis - Photometry	Tréguer and Le Corre 1975; Aminot and Kérouel 2007
	Nitrite	1980-2013 (monthly)	1980-2013 (monthly)	Flow analysis – Photometry	NF-EN ISO 26667		Flow analysis - Photometry	Tréguer and Le Corre 1975; Aminot and Kérouel 2007
	Ammonium	1980-2013 (monthly)	1980-2013 (monthly)	Manual - Photometry	NF-EN-ISO 11732	1997-2013 (bimonthly)	Manual - Photometry	Aminot and Chaussepied 1983
							Flow analysis - Fluorimetry	Aminot and Kérouel 2007
DIP (μmol L^{-1})	Phosphate	1990-2013* (monthly)	1990-2013* (monthly)	Flow analysis - Photometry	NF-EN ISO 6678	1997-2013 (bimonthly)	Manual - Photometry	Aminot and Chaussepied 1983
							Flow analysis - Photometry	Tréguer and Le Corre 1975; Aminot and Kérouel 2007
DSi (μmol L^{-1})	Silicate	2002-2013 (monthly)	2003-2013 (monthly)	Flow analysis - Photometry	NF-EN-ISO 16264	1997-2013 (bimonthly)	Manual - Photometry	Aminot and Chaussepied 1983
							Flow analysis - Photometry	Tréguer and Le Corre 1975; Aminot and Kérouel 2007
Chl *a* (μg L^{-1})		1980-2013 (monthly)	1980-2013 (monthly)	Photometry	NF T 90-117	1996-2013 (bimonthly)	Fluorimetry	Neveux and Panouse 1987; Aminot and Kérouel 2004
							Spectrophotometry	Aminot and Chaussepied 1983; Aminot and Kérouel 2004
Micro-phytoplankton counts (cells L^{-1})	Diatoms					1983-2013 (bimonthly)	Lugol fixed sample	Utermöhl 1958
	Dinoflagellates					1983-2013 (bimonthly)	Lugol fixed sample	Utermöhl 1958
River discharges (m^3 s^{-1})		1980-2013 (daily)	1980-2013 (daily)					

**Note: DIP measurements were less reliable from 1980 to 1989 in the Loire and in the Vilaine, and between 2009 and 2011 in the Vilaine only*

Table S2: Annual median values for nutrients and chlorophyll *a* concentrations measured in offshore waters of the Bay of Biscay. Minimum and maximum values are indicated in parentheses. Data numbers are given in brackets. Dataset was extracted from the ICES Oceanographic database for the period from 1995 to 2002 (http://ocean.ices.dk/HydChem/HydChem.aspx?plot=yes) at a geographical zone between 3-6 °W and 45-47 °N. For the period from 2010 to 2016, dataset was collected from the "PELGAS" oceanography expedition "PELGAS" (http://campagnes.flotteoceanographique.fr/series/18/). * Nitrate was measured as nitrate + nitrite. The sampling frequency was highly irregular, happening mainly in summer and for a short period only, thus preventing trend analysis

Year	Nitrate (µmol L^{-1})	Nitrite (µmol L^{-1})	Ammonium (µmol L^{-1})	DIP (µmol L^{-1})	DSi (µmol L^{-1})	Chl *a* (µg L^{-1})
1995	4.7 (3.4; 6.9) [21]	0.06 (0.03; 0.19) [39]		0.44 (0.05; 0.80) [55]	2.1 (0.50; 3.5) [48]	0.50 (0.30; 0.50) [15]
1996		0.05 (0.03; 0.14) [11]		0.43 (0.05; 0.90) [46]	2.8 (0.50; 6.6) [44]	
1997						
1998	5.9 (5.3; 6.6) [15]	0.04 (0.03; 0.16) [40]		0.37 (0.05; 0.96) [46]	2.2 (0.50; 4.6) [38]	
1999						
2000	2.5 (0.20; 6.9) [52]	0.05 (0.03; 0.14) [20]	0.20 (0.0; 1.00) [58]	0.19 (0.05; 0.63) [39]	1.80 (0.60; 4.7) [44]	0.60 (0.10; 2.3) [81]
2001	2.4 (0.0; 7.7) [75]	0.10 (0.03; 0.17) [39]	0.25 (0.0; 1.00) [76]	0.19 (0.05; 0.83) [52]	2.2 (0.80; 6.4) [66]	0.50 (0.10; 6.4) [44]
2002	3.0 (1.00; 5.4) [21]	0.11 (0.03; 0.15) [16]	0.10 (0.0; 0.80) [34]	0.25 (0.15; 0.87) [22]	1.60 (0.90; 5.3) [27]	0.60 (0.20; 1.60) [13]
2003						
2004						
2005						
2006						
2007						
2008						
2009						
2010	0.60 (0.01; 6.3) [54]*		0.66 (0.13; 4.31) [55]	0.02 (0.0; 0.30) [55]	0.85 (0.34; 2.2) (56)	1.16 (0.11; 10.1) [60]
2011	0.0 (0.0; 6.4) [53]*		0.0 (0.0; 0.26) [53]	0.04 (0.0; 0.13) [53]	0.62 (0.02; 6.7) [53]	0.43 (0.16; 2.8) [53]
2012	1.20 (0.50; 5.1) [55]*		0.13 (0.05; 2.4) [55]	0.11 (0.05; 0.35) [55]	1.30 (0.20; 4.8) [55]	
2013						
2014	0.90 (0.20; 18.6) [62]*		0.19 (0.05; 1.63) [62]	0.05 (0.05; 0.54) [62]	1.10 (0.40; 14.5) [62]	1.32 (0.28; 11.8) [65]
2015	0.50 (0.50; 12.9) [54]*		0.23 (0.05; 4.9) [54]	0.05 (0.05; 0.50) [54]	0.58 (0.05; 7.7) [54]	1.58 (0.24; 13.5) [63]
2016	0.50 (0.05; 18.5) [60]*		0.19 (0.05; 1.12) [60]	0.05 (0.05; 0.32) [60]	0.94 (0.40; 12.8) [60]	1.47 (0.41; 7.2) [94]

Nutrient and Chl a concentrations in the Bay of Biscay

Nutrient and Chl *a* in the Bay of Biscay are presented here for a comparison of nutrients sources to Vilaine Bay. For the period of 1995-2002, data were extracted from ICES Oceanographic database (http://ocean.ices.dk/HydChem/HydChem.aspx?plot=yes) at the geographical zone between 3-6° W and 45-47° N. These data are compiled with those from PELGAS surveys (http://campagnes.flotteoceanographique.fr/series/18/) for the period of 2010-2016 (see Doray et al., 2017). Dataset in the Bay of Biscay were not included for trend analysis due to the short periods and irregularity of sampling, occurring mainly in summer. Nutrients and Chl *a* in the Bay of Biscay were always lower than those measured in rivers and in VB and did not display any increase from 1995 to 2016.

Table S3: Statistical results of modified Mann-Kendal test performed on DLM trend components of dissolved inorganic nutrient ratios in rivers and in the VB for the common period 1997–2013. Changes were calculated as differences of the Sen's robust line between the beginning and the end of the period (17 years) if the test was significant at $p<0.05$. Values in parentheses are percentages of changes relative to the initial values of the Sen's robust line. Cells were left blank when tests were not applicable

Site/ parameters	Loire		Vilaine		Vilaine Bay	
	p	Change (%)	*p*	Change (%)	*p*	Change (%)
DIN:DIP	<0.001	+ 92 (85%)	<0.001	+ 410 (303%)	<0.001	+ 13 (72%)
DIN:DSi					0.009	+ 0.15 (21%)
DSi:DIP					<0.001	+ 15 (57%)

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Figure S1: Scatter plot of DIP concentrations between 1980 and 2013 in the Loire (a) and in the Vilaine (b). Note series of constant (repetitive) values for the period before 1990 in the Loire and between 2009 and 2011 in the Vilaine (see Section 2.3.1)

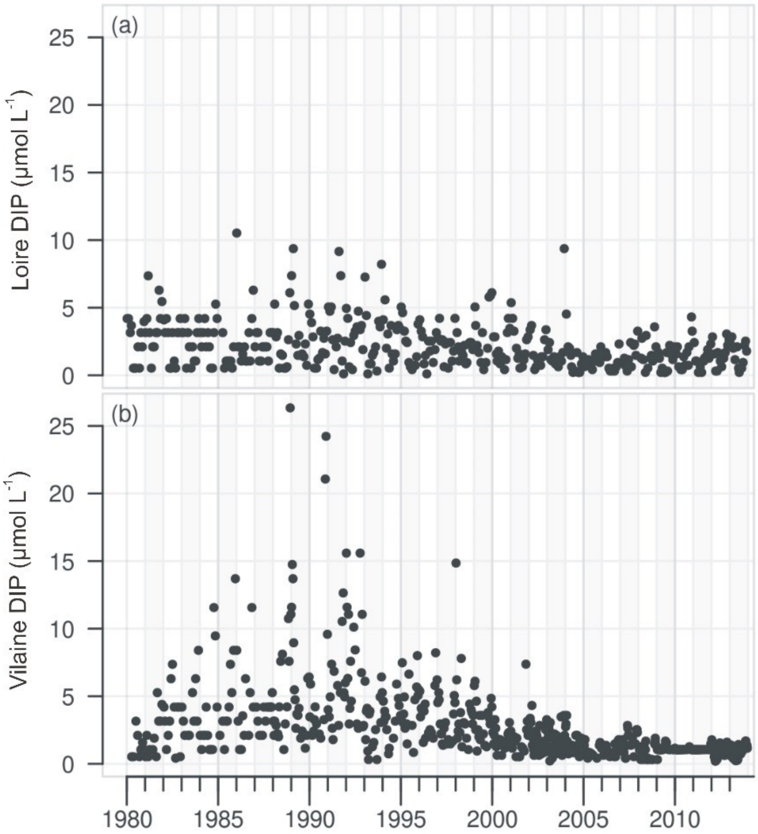

Figure S2: Long-term trend and seasonality of river discharges in the Vilaine (a, b). Dark grey lines represent DLM trends. Shaded areas indicate the 90 % confidence interval. Each dot in the trend plot (left) represents an observed value, those in the seasonality plot (right) represent values estimated 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 indicates the beginning of the longest common period for all studied variables in rivers and in the VB (1997-2013)

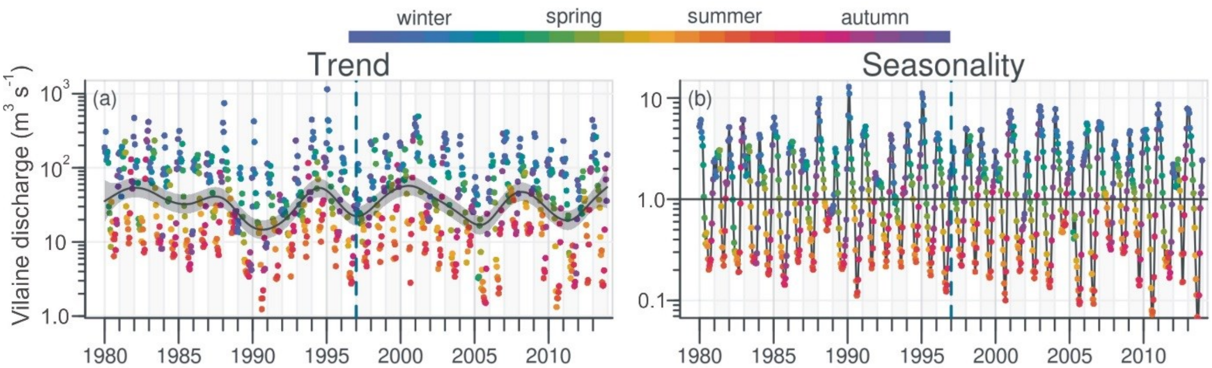

Figure S3: Long-term trend and seasonality of DIP in the Vilaine (a, b) and DIP loads from the Vilaine (c, d). Note: analytical problems of DIP measurements between 2009 and 2011 (see Section 2.3.1). See Fig. S2 for details

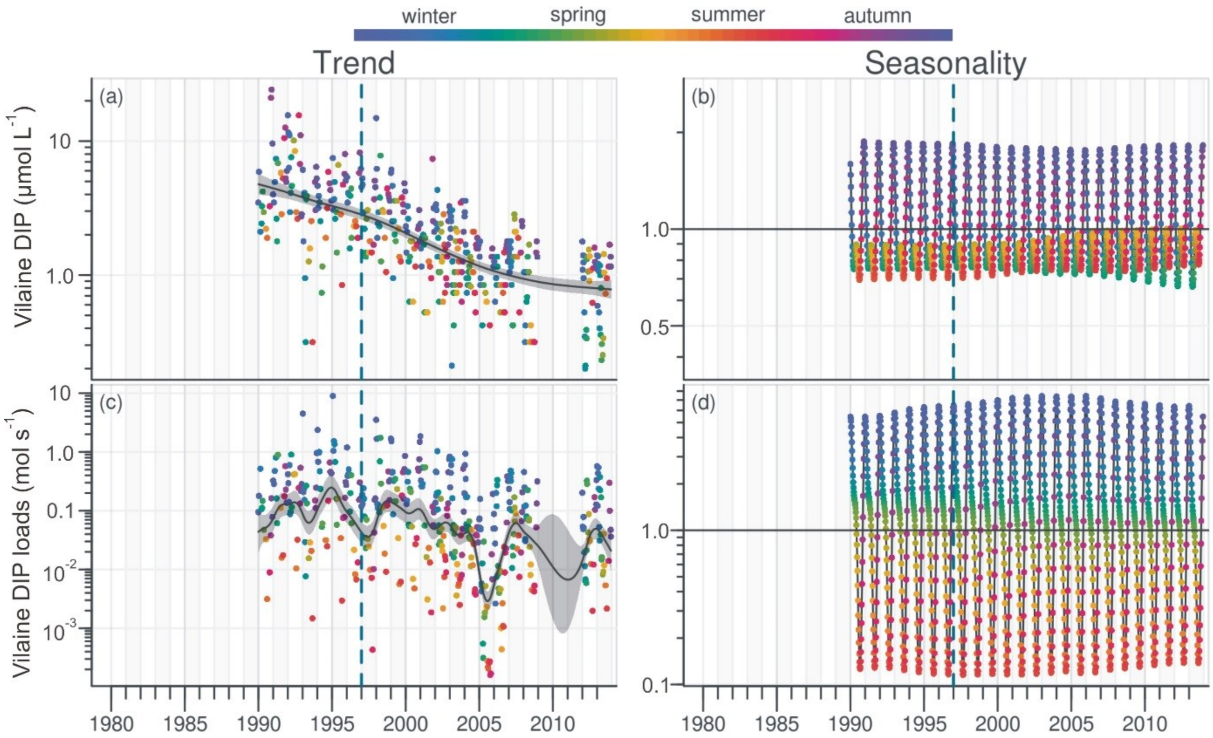

Figure S4: Long-term trend and seasonality of DIN in the Vilaine (a, b) and DIN loads from the Vilaine (c, d). Black dots represent data considered as outliers (see Section 2.3.2). See Fig. S2 for details

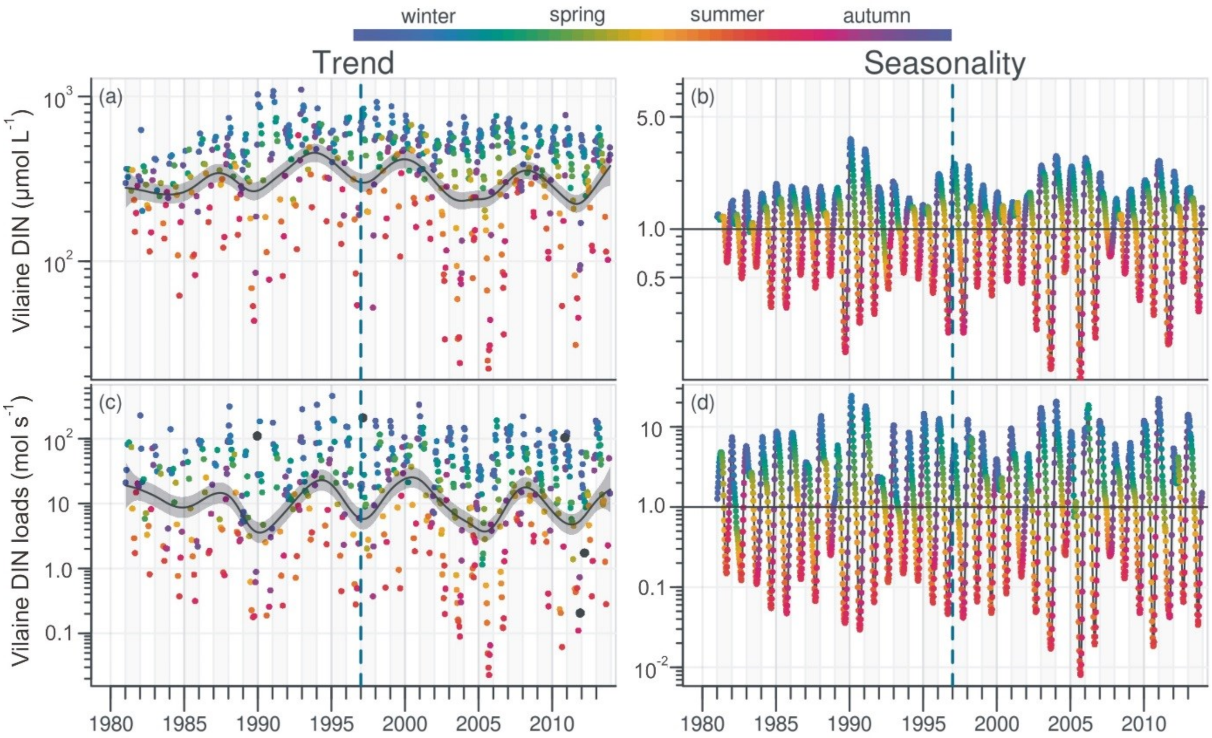

Figure S5: Long-term trend of DIN:DIP ratios in the Loire (a) and Vilaine (b) rivers. Black dots represent data considered as outliers (see Section 2.3.2). Dashed horizontal black line corresponds to theoretical value for DIN:DIP of 16. See Fig. S2 for details

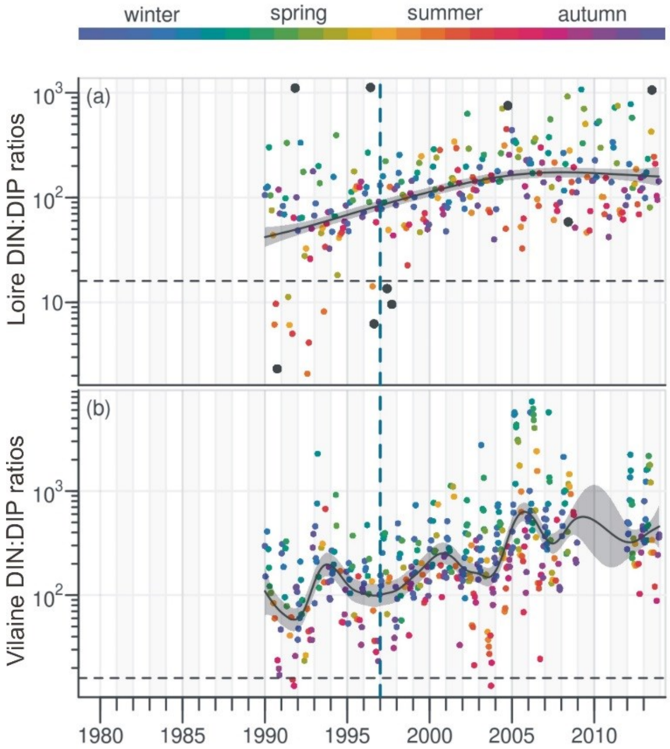

Figure S6: Long-term trend and seasonality of Chl *a* in the Vilaine (a, b). See Fig. S2 for details

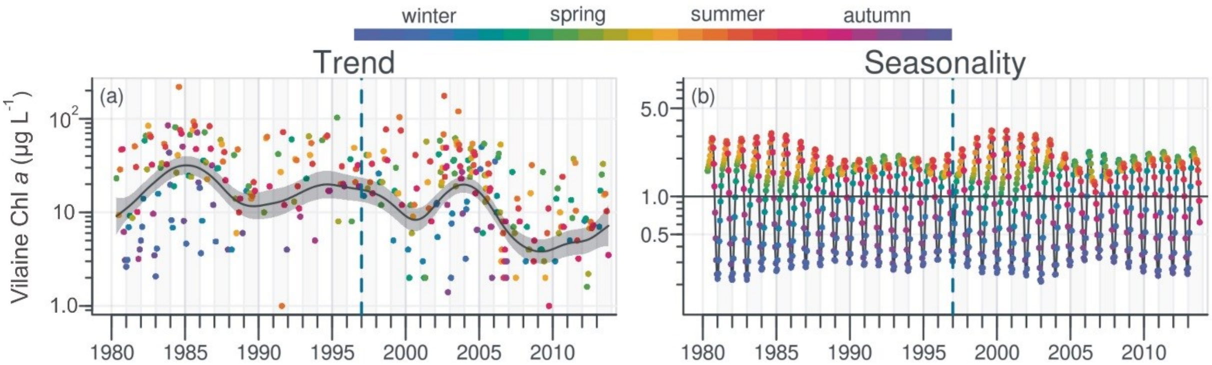

Figure S7: Long-term trend of DIN:DIP (a), DIN:DSi (b), and DSi:DIP (c) ratios in the VB. Black dots represent data considered as outliers (see Section 2.3.2). Dashed horizontal black line corresponds to theoretical value for DIN:DIP, DIN:DSi, and DSi:DIP respectively of 16, 1 and 16. See Fig. S2 for details