Supplementary materials

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

Widya Ratmaya[1*], Dominique Soudant[2], Jordy Salmon-Monviola[3], Nathalie Cochennec-Laureau[1], Evelyne Goubert[4], Françoise Andrieux-Loyer[5], Laurent Barillé[6], Philippe Souchu[1]

[1] Ifremer – LER MPL, Rue de l'Ile d'Yeu, BP 21105, 44311 Nantes Cedex 03, France

[2] Ifremer, VIGIES, Rue de l'Ile d'Yeu, BP 21105, 44311 Nantes Cedex 03, France

[3] INRA/Agrocampus Ouest – UMR1069 Sol Agro et Hydrosystème Spatialisation, 35000 Rennes, France

[4] Université de Bretagne Sud – GMGL, UMR CNRS 6538 DO, Campus Tohannic, 56000 Vannes, France

[5] Ifremer – DYNECO PELAGOS, ZI Pointe du Diable, 29280 Plouzané, France

[6] Université de Nantes, Mer Molécules Santé EA 2160, Faculté des Sciences et des Techniques, BP 92208, 44322 Nantes cedex 3, France

* Email : widya.ratmaya@ifremer.fr

Laboratoire Environnement Ressource Morbihan – Pays de la Loire, Centre Ifremer de Nantes, Rue de l'Ile d'Yeu, BP 21105, 44311 Nantes Cedex 03, France.

Table S1: Available datasets for all sites of this study

Sites/ Parameter		Loire	Vilaine	Method	References	Vilaine Bay	Method	References
DIN ($\mu mol\ L^{-1}$)	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 ($\mu mol\ L^{-1}$)	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 ($\mu mol\ L^{-1}$)	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* ($\mu g\ L^{-1}$)		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 1995 to 2002, 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 ($\mu mol\ L^{-1}$)	Nitrite ($\mu mol\ L^{-1}$)	Ammonium ($\mu mol\ L^{-1}$)	DIP ($\mu mol\ L^{-1}$)	DSi ($\mu mol\ L^{-1}$)	Chl *a* ($\mu g\ L^{-1}$)
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 a 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 2003-2016 (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 (Table A1) and did not display any increase from 1995 to 2016.

References

Aminot, A., and M. Chaussepied. 1983. *Manuel des analyses chimiques en milieu marin (in French)*. Paris, France: CNEXO.

Aminot, A., and R. Kérouel. 2004. *Hydrologie des écosystèmes marins: paramètres et analyses (in French)*. Plouzané, France: Ifremer.

Aminot, A., and R. Kérouel. 2007. *Dosage automatique des nutriments dans les eaux marines: méthodes en flux continu (in French)*. Plouzané, France: Ed. Ifremer.

Doray, M., Petitgas, P., Romagnan, J. B., Huret, M., Duhamel, E., Dupuy, C., Spitz, J., Authier, M., Sanchez, F., Berger, L., Dorémus, G., Bourriau, P., Grellier, P., and Massé, J.: The PELGAS survey: Ship-based integrated monitoring of the Bay of Biscay pelagic ecosystem, Prog Oceanogr, https://dx.doi.org/10.1016/j.pocean.2017.09.015, 2017.

Doray, M., Duhamel, E., Huret, M., Petitgas, P., Masse, J.: PELGAS, http://dx.doi.org/10.18142/18, 2000.

Neveux, J., and M. Panouse. 1987. Spectrofluorometric determination of chlorophylls and pheophytins. *Archiv Fur Hydrobiologie* 109: 567-581.

Tréguer, P., and P. Le Corre. 1975. *Manuel d'analyse des sels nutritifs dans l'eau de mer: Utilisation de l'autoanalyzer II Technicon R (in French)*. Brest, France: Université de Bretagne Occidentale.

Utermöhl, H. 1958. Zur Vervollkommnung der quantitativen Phytoplankton-Methodik. *Mitteilungen Internationale Vereinigung Theoretische und Angewandte Limnologie* 9: 1-38.

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.)

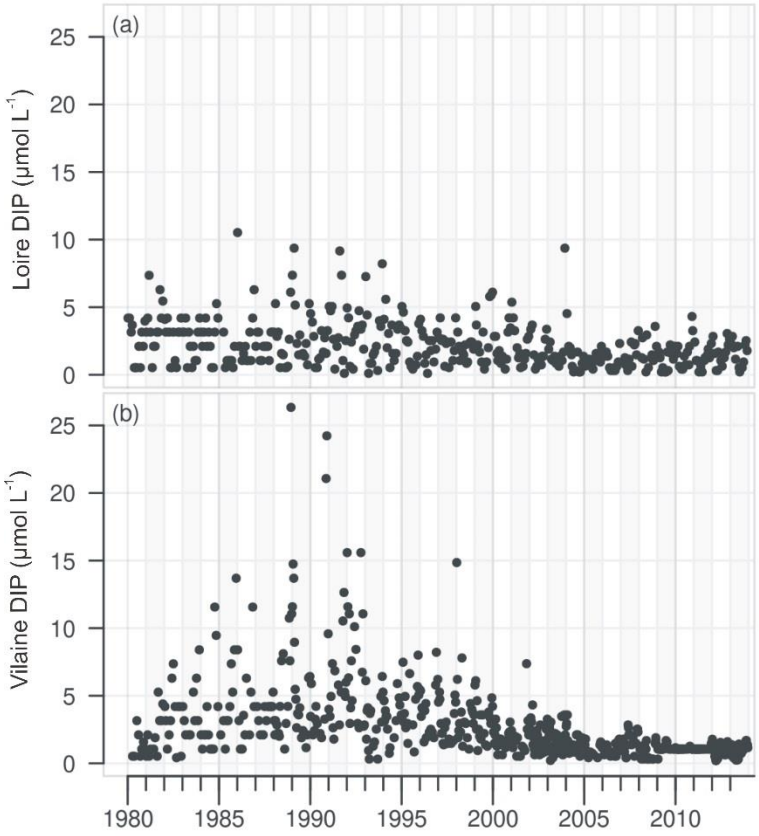

Figure S2: Long-term trends and seasonality of river discharges in the Vilaine (a, b). Dark grey lines represent DLM trends. Shaded areas indicate the 95 % confidence interval. Each dot in the trend plot (left) represents an observed value, those in the seasonality plot (right) represent values estimated by the model. Dashed vertical blue line corresponds to the period for MK test (1997-2013)

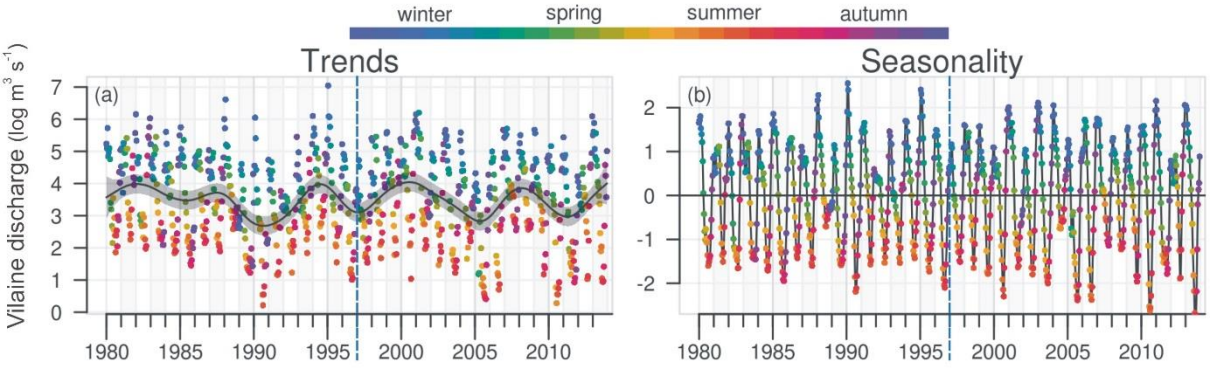

Figure S3: Long-term trends and seasonality of DIP in the Vilaine (a, b) and DIP loads from the Vilaine (c, d). Note: analytical problems of DIP measurements between 2009 and 2011 (see Section 2.4.). See Fig. S2 for details

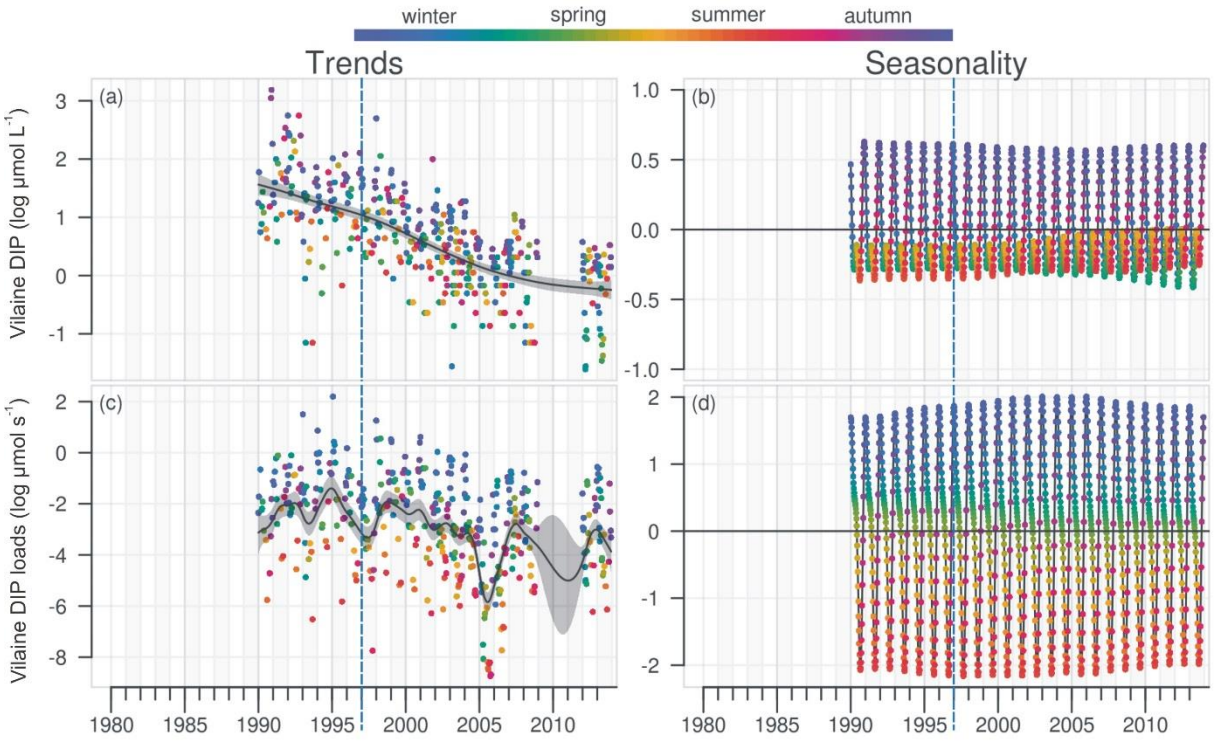

Figure S4: Long-term trends and seasonality of DIN in the Vilaine (a, b) and DIN loads from the Vilaine (c, d). Black dots represent data considered as outliers (see Section 2.4.). See Fig. S2 for details

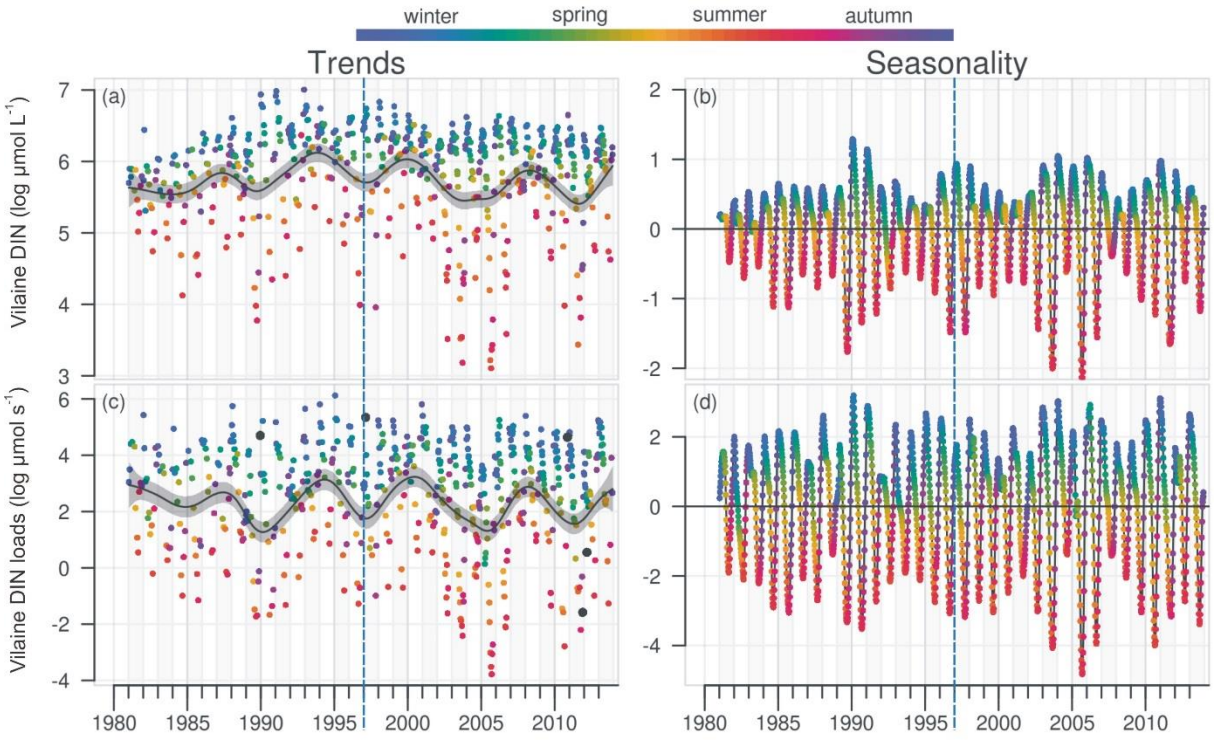

Figure S5: Long-term trends of DIN:DIP ratios in the Loire (a) and Vilaine (b) rivers. Black dots represent data considered as outliers (see Section 2.4.). See Fig. S2 for details

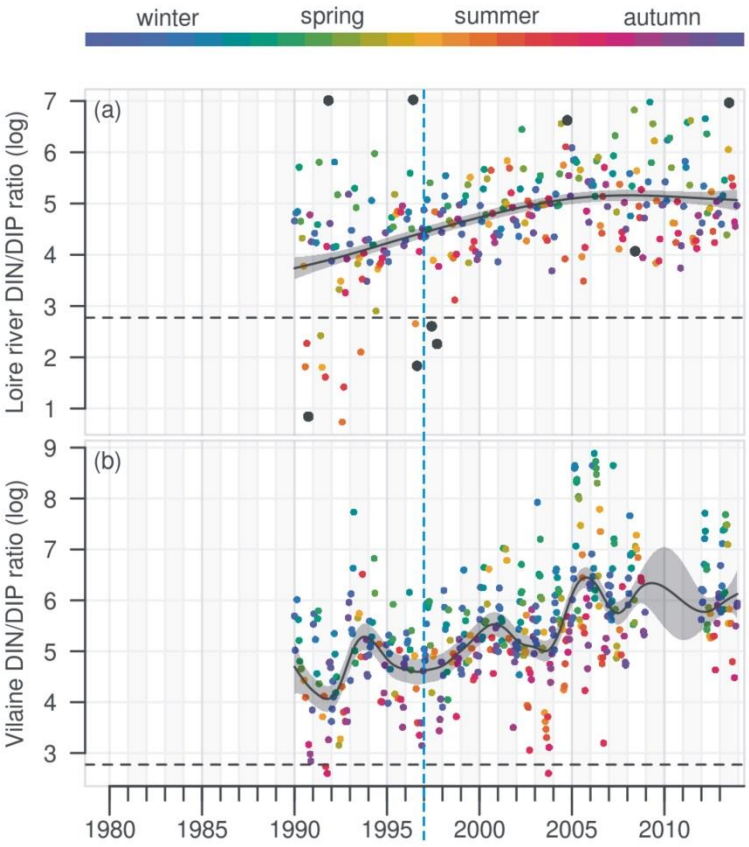

Figure S6: Long-term trends and seasonality of Chl *a* in the Vilaine (a, b). See Fig. S2 for details

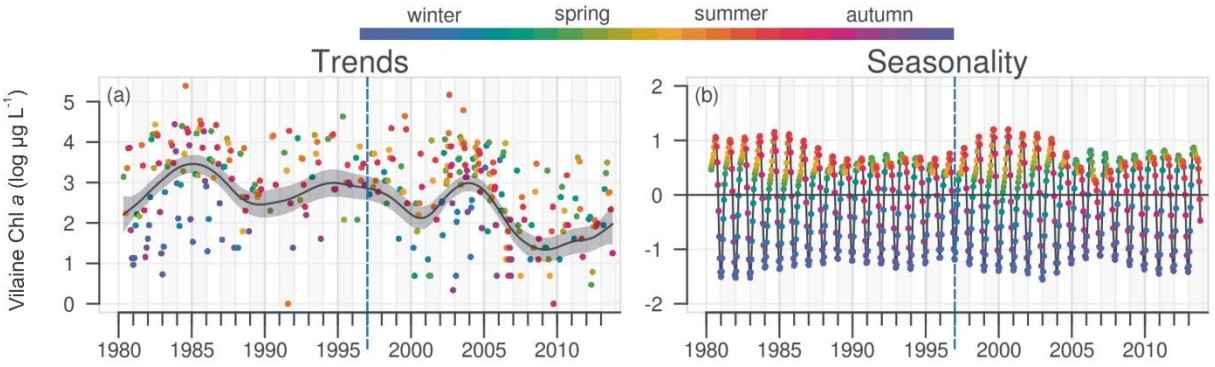

Figure S7: Long-term trends of DIN:DIP (a), DIN:DSi (b), and DSi:DIP (c) ratios in Vilaine Bay. Black dots represent data considered as outliers (see Section 2.4.). See Fig. S2 for details