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# Early evaluation of coastal nutrient over-enrichment: New procedures and indicators

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A B S T R A C T

Recent studies have provided compelling evidence for an accelerated anthropogenic impact on coastal systems, resulting in intense inputs of materials and nutrients from the continent. This has led scientists and policymakers to encourage the implementation of monitoring programmes, which have resulted in the multiplicity of datasets. However surprisingly, only a few attempts have been made to couple observations with statistical and mathematical tools to detect, as soon as the data become available perturbations in coastal systems. Here, we propose new mathematical procedures to evaluate the state of a system, based on the building of relative reference state and indicators of nutrient over-enrichment. The techniques were tested in some French coastal systems using data from the programme SOMLIT. Applied to this dataset, the multivariate procedures rapidly identified and evaluated anthropogenic nutrient anomalies from the continent on three sites (Wimereux, Roscoff and Villefranche-sur-Mer) from 1997 onwards.



## 1. Introduction

The anthropogenic use of both freshwater and land systems has greatly increased over time (Conley, 2000), altering marine systems at an unprecedented rate on a variety of temporal and spatial scales (Dobson et al., 1997; Halpern et al., 2008). These perturbations have resulted in an increase in nutrient inputs from the continent to the sea, which have altered both the structure and the functioning of coastal systems with potential implications for some natural biogeochemical cycles (Vitousek et al., 1997a; Buddemeier et al., 2002). As a consequence, dramatic disruptions of coastal systems have been observed (Cloern, 2001; Selman et al., 2008) leading sometimes to toxic or harmful algal blooms (Anderson et al., 2002), depletion or increase in dissolved oxygen (Diaz, 2001; Paerl, 2006; Vaquer-Sunyer and Duarte, 2008) and an alteration in species diversity (Richardson et al., 1997).

While anthropogenic influences on terrestrial ecosystems are perhaps more easily quantified and documented (e.g. Vitousek et al., 1997b; Smith et al., 1999; Vorosmarty and Sahagian, 2000), the relationships between nutrient inputs, eutrophication, and hypoxia/anoxia dynamics in marine ecosystems (Cloern, 2001; Halpern et al., 2008) remain difficult to understand (Conley, 2000; Mysterud et al., 2001; Paerl, 2006). The worldwide accumulation of evidence of the alteration in coastal systems led scientists and policymakers to encourage the implementation of monitoring programmes. However, once a coastal marine region is monitored, there is also a need to develop indicators to measure the changing state of the system (Balmford et al., 2003). Indicators provide insights on the state and dynamics of the environment (Niemeijer, 2002). However, creating some indicators remains challenging. For example, it remains difficult to create indicators that separate the influence of climate change from the more direct impact of human activities (Borja et al., 2008). The degree of anthropogenic perturbation of a coastal system should be compared with a site in which only natural conditions are a source of variability (i.e. a pristine, unpolluted, or anthropogenically undisturbed state; Hughes, 1994; Davies and Jackson, 2006). However, such reference states rarely exist (Goberville et al., 2011).

The main objectives of this study were (1) to propose new numerical procedures that enable the identification and the quantification, as soon as data become available, of a potential anthropogenic perturbation and (2) to offer new indicators of human nutrient over-enrichment to better monitoring a coastal system. Our procedures were applied on three French sites

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monitored by the programme SOMLIT (Wimereux, Roscoff and Villefranche-sur-Mer) from 1997 onwards.

## 2. Environmental data

Our analyses were based on data collected at least twice a month by the French sampling programme SOMLIT. This programme has coordinated activities of a number of marine stations along the French coasts. It currently comprises seven marine stations at twelve sampling sites and monitors all seas surrounding France such as the English Channel, the Atlantic Ocean and the Mediterranean Sea. Coastal systems monitored by this programme have distinctive hydro-climatic characteristics ranging from the weak-tidal Mediterranean Sea (e.g. Villefranche-sur-Mer) to the mega-tidal English Channel (e.g. Wimereux) (Fig. 1). More details on the monitoring programme are available at http://www.insu.cnrs.fr/co/services-nationaux-labellises/ocean-atmosphere-climat/surveillance-ocean/.

On the seven stations and 12 sites monitored by SOMLIT, we focussed our analyses on the three stations for which both inshore and offshore information were available: Wimereux, Roscoff and Villefranche-sur-Mer (Fig. 1 and Table 1). In Villefranche-sur-Mer, the offshore site was not a component of the programme SOMLIT but was part of the programme DYFAMED ('DYnamique des Flux Atmosphériques en MEDiterranée'), a long-term time-series investigation performed in the context of the programme JGOFS ('Joint Global Ocean Flux Study'). The DYFAMED site was located in the central zone of the Ligurian Sea (Fig. 1), and was a homogeneous system too remote to be influenced by direct coastal inputs by the rivers. From January 1991 to July 2007, monthly cruises were conducted and most of the JGOFS core parameters recorded (Knap et al., 1996). All data are available through the DYFAMED Observatory data base (http://www.obs-vlfr.fr/sodyf/).

In this study, we focussed on four parameters: Sea Surface Temperature (SST), nitrate, phosphate and silicate concentrations for all sites. All data were collected at subsurface. Because of the dependency on meteorological conditions during sampling, there were some missing data in the matrices (Marty et al., 2002; Goberville et al., 2010).

## 3. Methods

Previous results have shown that the concentration of both nitrate and phosphate along the French Coast was highly influenced by the climatic regime (Goberville et al., 2010). The relationship was detected at a seasonal scale. The basic pattern is that winter is the season during which the concentration in nutrient increases whereas summer is characterised by a reduction in nutrient (e.g. Gentilhomme and Lizon, 1997). In addition to this well-known feature, year-to-year changes in nutrient concentration were also strongly affected by both SST and precipitation, which reflected large-scale variability in atmospheric circulation (Goberville et al., 2010). Interaction occurred between climate and human activities, the anthropogenic nutrient over-enrichment being either amplified by increased precipitation or attenuated by rising temperature and conversely.

### 3.1. Analyses 1: indicators of anthropogenic nutrient concentration

To remove the confounding effect of climate, indicators of anthropogenic nutrient concentration were created by calculating the residuals of both nitrate and phosphate concentrations after having applied a Multiple Linear Regression (MLR) using as explanatory variables both SST and silicates. The MLR was performed on data merging information on inshore and offshore sampling site for three stations: Wimereux, Roscoff and Villefranche-sur-Mer. Prior to the analysis, data were averaged per month. SST was selected because it reflects a major influence of climate on biological and chemical systems (Cloern, 1996). Silicates were chosen because the predominant source of dissolved silica is the natural weathering of silicate minerals (Tréguer et al., 1995). At a given site, their concentration increases with precipitation and river runoff, another important influence of climate (Labat et al., 2004). Moreover, silicate concentrations are not directly influenced by human activities

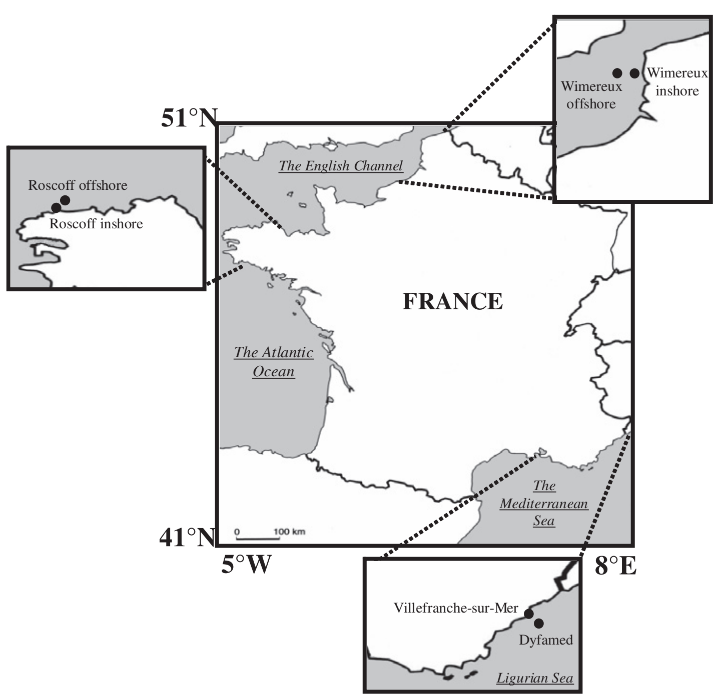


**Fig. 1.** Location of inshore and offshore sites of the three stations examined in this study.

**Table 1**
Main characteristics of the sites Wimereux, Roscoff and Villefranche-sur-Mer. For location of sites see Fig. 1.

| | Wimereux | | Roscoff | | Villefranche-sur-Mer | |
|---|---|---|---|---|---|---|
| Site | Inshore | Offshore | Inshore (Estacade) | Offshore (Astan) | Inshore (SOMLIT site) | Offshore (DYFAMED site) |
| Longitude | 1° 31' 17 E | 1° 24' 60 E | 3° 58' 58 W | 3° 56' 15 W | 7° 19' 00 E | 7° 52' 00 E |
| Latitude | 50° 40' 75 N | 50° 40' 75 N | 48° 43' 56 N | 48° 46' 40 N | 43° 41' 00 N | 43° 25' 00 N |
| Time series | November 1997–December 2009 | November 1997–December 2009 | January 1997–December 2009 | February 2000–December 2009 | January 1993–December 2009 | January 1991–July 2007 |
| Tide condition | HT–ST Coef 95±25 | HT–ST Coef 95±25 | HT–NT | HT–NT | – | – |
| Distance to coast (in m) | 1850 | 9260 | 500 | 3500 | 500 | 5200 |
| Water depth (in m) | 26 | 53 | 3 | 60 | 95 | 2350 |
| Marling (in m) | S.T. mean (Coef: 95): 7.7 | S.T. mean (Coef: 95): 7.7 | S.T. mean (Coef: 95): 7.5 | S.T. mean (Coef: 95): 7.5 | Barometric: 0.2 | Barometric: 0.2 |
| River runoff influence | Somme and Liane Rivers | Negligible | Penzé River and stormwater | Negligible | Negligible | Negligible |

HT: High Tides; ST: Spring Tides; NT: Neap Tides; Coef: Tidal Coefficient.

contrary to nitrate and phosphate concentrations (Muylaert et al., 2009). Therefore for each station (Wimereux, Roscoff and Villefranche-sur-Mer), two MLRs were applied with nitrates (first MLR) and phosphates (second MLR) as the predictive variable and both SST and silicates as explanatory variables (first and second MLR; Fig. 2 and Supplementary Figs. 1 and 2). The general model applied was therefore as follows (Sokal and Rohlf, 1995):

$$\hat{Y} = a + b.x_1 + c.x_2 \quad (1)$$

where the dependent variable $\hat{Y}$ (also called prediction of the model, i.e. nitrate or phosphate concentrations) was a function of the two explanatory variables $x_1$ (SST) and $x_2$ (silicate concentrations), $a$ the intercept, $b$ and $c$ the partial regression coefficients. Residuals of nitrate and phosphate concentrations were then obtained by calculating the difference between the observed and predicted value for a given observation in the time series and for each station. A total of six MLRs were thereby applied, giving an indication of both nitrate and phosphate anthropogenic concentration for each of the three stations under investigation.

Partial correlation coefficients were examined to evaluate the influence of each variable (SST and silicates) on nitrate and phosphate concentrations. The (first-order) partial correlation coefficient between variable $y_1$ (nitrate concentrations) or $y_2$ (phosphate concentrations) and the two explanatory variables $x_1$ (SST) and $x_2$ (silicates) was assessed while keeping either $x_1$ or $x_2$ linearly constant. For example, the partial correlation coefficient between the two variables $y_1$ and $x_1$ with $x_2$ held constant was calculated as follows:

$$r_{y_1,x_1.x_2} = \frac{r_{y_1,x_1} - r_{y_1,x_2} r_{x_1,x_2}}{\sqrt{\left(1 - r^2_{y_1,x_2}\right)\left(1 - r^2_{x_1,x_2}\right)}} \quad (2)$$

where $r_{y_1,x_1}, r_{y_1,x_2}$ and $r_{x_1,x_2}$ is the simple correlation coefficient between variables $y_1$ and $x_1$, $y_1$ and $x_2$ and $x_1$ and $x_2$, respectively. The coefficient of multiple determination $R^2$ was tested by analysis of variance (Sokal and Rohlf, 1995). An analysis of variance was performed to test the significance of the addition of each independent variable in the regression. Because of the limited number of variables included in the analysis, the multiple regression was performed using both the forward selection and the backward elimination procedures (Legendre and Legendre, 1998).

### 3.2. Analyses 2: mapping of the system state and both spatial and temporal comparisons

To monitor a system, it is important to evaluate its current state and to compare it to some reference values incorporating some measure of natural variability (García et al., 2010). We therefore needed what can be called reference conditions or a reference state (Niemi et al., 2004). Establishing a reference state, i.e. quantifying the actual state of an ecosystem and its natural variability, is also a prerequisite to better identify potential effects of disturbances (Binet, 1997). However, the notion of reference state remains debated, mainly because it should not be considered as a pristine state (Niemeijer, 2002; Niemi et al., 2004). We therefore chose to use the term "relative reference state", meaning here a system that is not substantially affected by human activities. This notion of relative reference state was attended to be an operational concept as it is obvious that it is to some extent affected by human activities. Inshore sites (potentially strongly affected by continental human influence) were thereby compared to the less affected offshore sites (considered here to be the relative reference state) to evaluate the direct human nutrient over-enrichment on the three selected coastal systems (inshore sites). To do so, we constructed two data matrices (one for inshore sites and the second for offshore sites) for each of the three stations (Wimereux, Roscoff and Villefranche-sur-Mer). The resulting tables were composed of the two indicators of both nitrate and phosphate anthropogenic concentration (see Analyses 1).

While for the inshore sites data were kept unchanged, this was not so for each data matrix of all offshore sites. A cubic or linear interpolation procedure (de Boor, 1978) was performed for each parameter to fill in some missing values. The interpolation was used to establish more robust training sets to create the offshore reference states for the three stations. The two interpolation procedures were due to the fact that when a large number of data were missing, applying only linear or cubic functions returned some unrealistic results. Potential biases related to the use of the interpolation procedures were checked by the use of boxplots for each parameter.

A total of 36 reference matrices was therefore defined for each month and each studied station (12 months × 3 offshore sites = 36 reference matrices). They all represented the relative reference states (i.e. the offshore sites) against which inshore sites were compared. To quantify the degree to which an observation (i.e. a given year) of the inshore site belonged to the relative reference state, we applied a technique based on a procedure recently proposed by Beaugrand et al. (2011) and Goberville et al. (2011). This technique calculates the probability of an observation to belong to the relative reference state.

The procedure had two main steps. First, for a given month and a studied station, the generalised Mahalanobis distance was calculated between each inshore observation (matrix $X_{n,p}$; $n$ observations and $p$ variables) to be tested against the relative reference

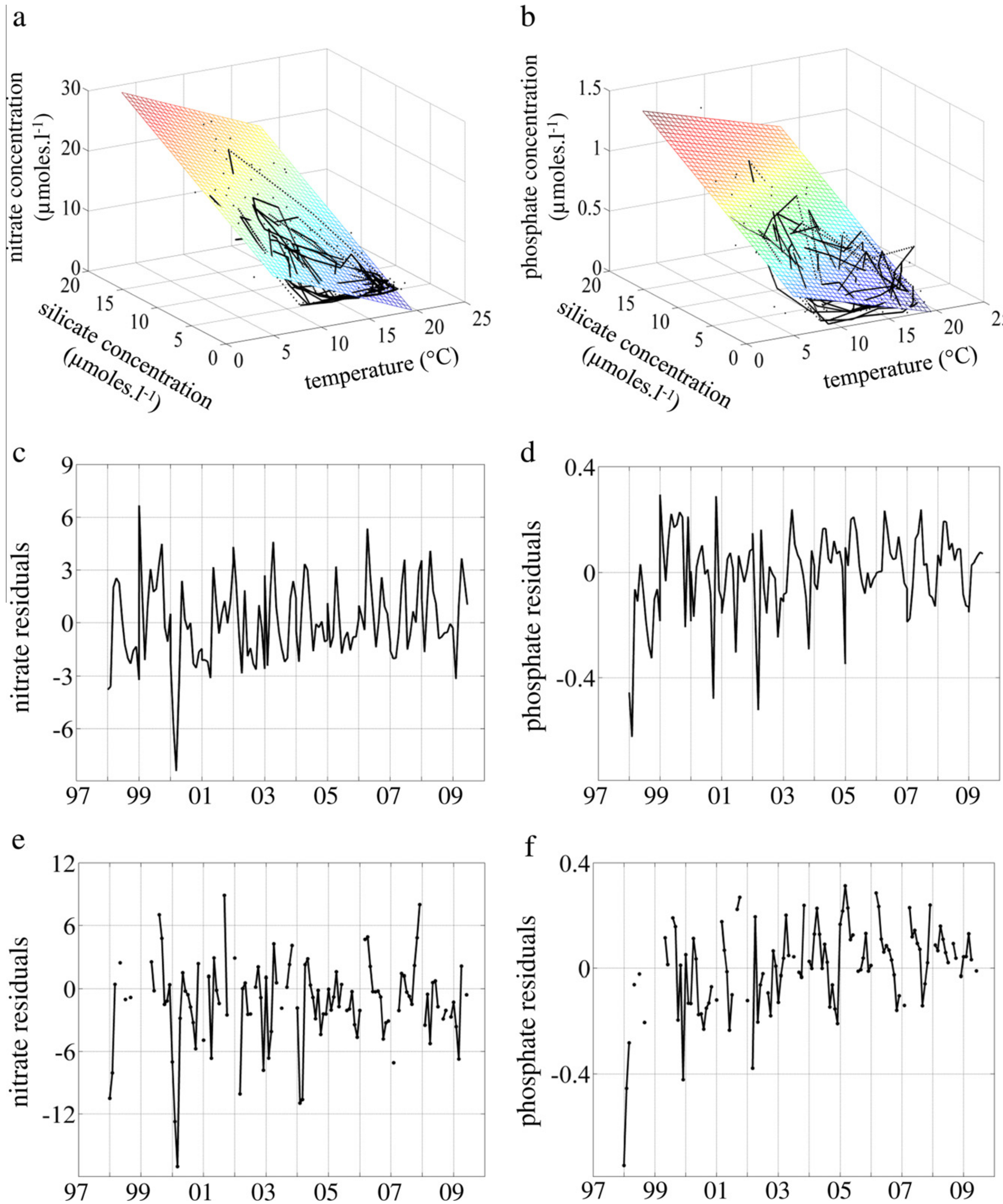


**Fig. 2.** Removing of the climatic influence in the time series of nitrate and phosphate in Wimereux. Multiple linear regressions using (a) nitrate concentration or (b) phosphate concentration as response variables and temperature and silicate concentration as explanatory variables. Year-to-year changes in nitrate anthropogenic concentration (c) offshore and (e) inshore. Year-to-year changes in phosphate anthropogenic concentration (d) offshore and (f) inshore.

state (i.e. all offshore samplings; matrix $Y_{m,p}$; $m$ observations and $p$ variables). We used the Mahalanobis generalised distance because it is independent of the scales of the descriptors and because it takes into consideration the covariance (or the correlation) among descriptors (Ibañez, 1981). To calculate the Mahalanobis generalised distance between each observation of the environment $y_i$ ($1 \leqslant i \leqslant m$) and all observations of the reference matrix $x_j$ ($1 \leqslant j \leqslant n$), we used a particular form of the generalised distance, giving the distance between a given observation and the centroid of a unique group (Ibañez, 1981):

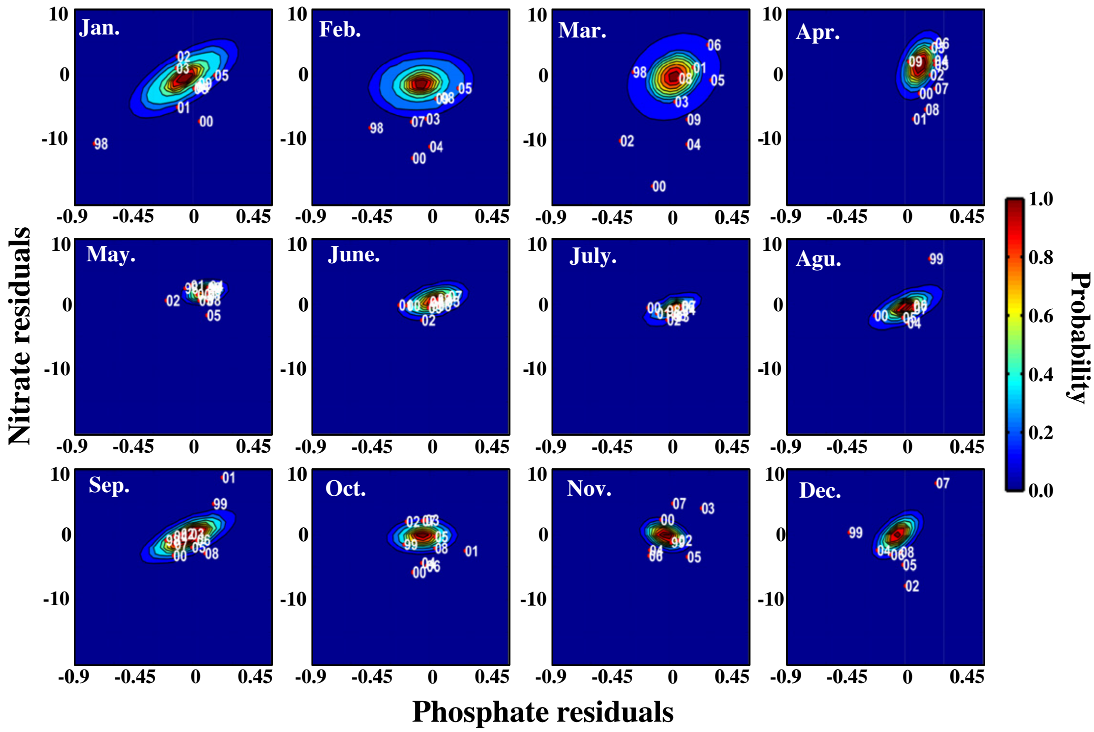


**Fig. 3.** Mapping of relative reference state in Wimereux as a function of both nitrate and phosphate anthropogenic concentration (residuals of the multiregression analysis; see Fig. 2) from January to December. The colour represents the probability that an observation belongs to the relative reference state. Each year from 1997 to 2009 was superimposed on the Euclidean space for the given month. The distance between a given year and the centroid of high probabilities was directly given by the probability of the year ($d = 1 - p$; see methods). Some years might be missing due to an absence of sampling because of adverse meteorological conditions.

$$D^2_{x_i,Y} = z'R^{-1}z \tag{3}$$

With $R_{p,p}$ the correlation matrix of table $Y_{m,p}$, $z$ the vector of the differences (of length $p$) between the values of the $p$ variables of observation $x_i$ and the average values of the $p$ variables of the offshore reference site contained in table $Y_{m,p}$. Then, the average observed distance $\varepsilon_0$ was calculated as follows:

$$\varepsilon_0 = \frac{\sum_{1=1}^{n} D_i}{n} \tag{4}$$

With $n$, the total number of Mahalanobis generalised distances $D_i$, equal to the number of inshore observations in $X$ (Beaugrand et al., 2011).

Second, the distance is tested by a simplified version of the Multiple Response Permutation Procedure (MRPP, Mielke et al., 1981; Beaugrand and Helaouët, 2008) performed on each offshore data set (Wimereux, Roscoff and Villefranche-sur-Mer). Mathematically, the simplified MRPP tests whether one observation belongs to a group of (reference) observations (Beaugrand and Helaouët, 2008). The number of maximum permutations is equal to $n$. After each permutation, the mean Mahalanobis generalised distance $\varepsilon_s$ is recalculated, with $1 \leqslant s \leqslant n$. A probability (called 'proM' latter in the text) can be assessed by looking at the number of times a simulated mean Mahalanobis generalised distance is found to be superior or equal to the observed mean Mahalanobis generalised distance between the observation and the reference matrix $X$ (Beaugrand et al., 2011).

$$\text{proM} = \frac{q_{\varepsilon_s \geq \varepsilon_0}}{n} \tag{5}$$

where the probability proM is the number of times the simulated mean Mahalanobis generalised distance was found superior or equal to the observed mean distance. When proM = 1, the observation statistically belongs to the centroïd of the relative reference state. When proM = 0, the observation has conditions in both nitrate and phosphate anthropogenic concentrations outside the relative reference state. Our procedure can detect all types of anomalies because the scale is determined by the probability level that ranges from 0 to 1. Applying the procedure to each observation of $Y_{m,p}$ leads to a matrix $P_{m,1}$ of probability.

The position of the offshore reference state was indicated in this space by high probabilities (e.g. see Fig. 3). The probability of each year of the period 1997–2009 for Wimereux and Roscoff and 1993–2009 for Villefranche-sur-Mer was calculated for a given month (from January to December) by calculating the generalised Mahalanobis distance and its probability as shown above. These probabilities were then superimposed on the 2-D Euclidean space represented by the indicators for both nitrate and phosphate anthropogenic concentrations (Figs. 3 and 4; Supplementary Figs. 3 and 4).

### 3.3. Analyses 3: year-to-year changes in the human nutrient over-enrichment

The new indicators proposed here enabled us to evaluate the state of the three coastal systems, focusing on both the rapid detection of anomalies and the intensity of human nutrient over-enrichment. The distance $d$ between a given year and the centre of high probabilities characterising the relative reference state was calculated as follows:

$$d = 1 - p \tag{6}$$

With $p$ the probability of a given year (see Analyses 2), the distance $d$ reflected the human nutrient over-enrichment of the inshore site (Figs. 5 and 6).

The 2-D Euclidean space centred on (0, 0) was defined with $x$ representing the residuals in phosphates and $y$ representing the residuals in nitrates (see Fig. 4). According to the indicators of both nitrate and phosphate anthropogenic concentrations, four cases

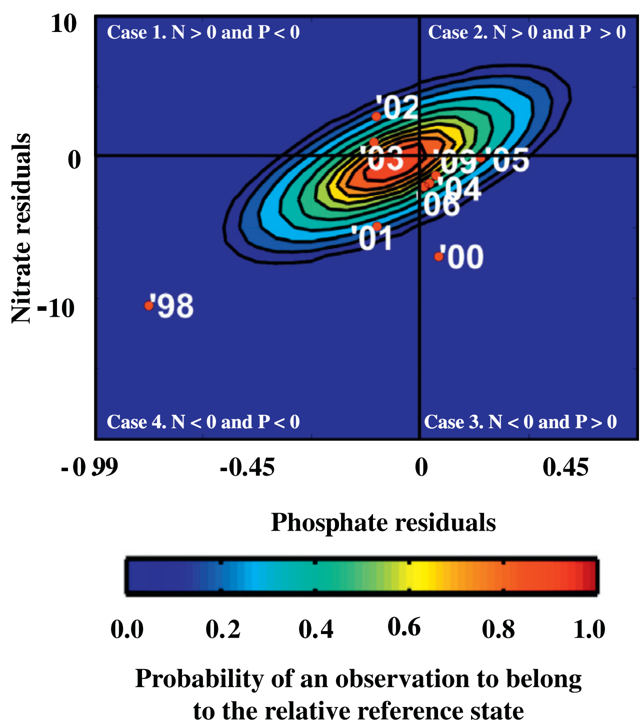


**Fig. 4.** Close examination of perturbations in both nitrate and phosphate in Wimereux in January (see the contour diagram performed for January in Fig. 3). Four cases were examined. Case 1: indicators for situations in which nitrate anthropogenic concentration was strictly superior to 0 and indicators in phosphate anthropogenic concentration strictly inferior to 0; Case 2: indicators for situations in which both nitrate and phosphate anthropogenic concentration are strictly superior to 0; Case 3: indicators for situations in which nitrate anthropogenic concentration are strictly inferior to 0 and indicators in situations when phosphate anthropogenic concentration are strictly superior to 0; Case 4: indicators for situations in which both nitrate and phosphate anthropogenic concentration are strictly inferior to 0. The colour represents the probability that an observation belongs to the relative reference state. Each year from 1997 to 2009 was superimposed on the Euclidean space for the given month. The distance between a given year and the centroid of high probabilities was directly given by the probability of the year ($d = 1 - p$; see methods). Some years might be missing due to an absence of sampling because of adverse meteorological conditions.

were examined (Fig. 4, Cases 1–4). The first case was characterised by an indicator of nitrate concentration >0 and an indicator of phosphate concentration <0 (Fig. 4, Case 1). The second case was characterised by an indicator of nitrate concentration >0 and an indicator of phosphate concentration >0 (Fig. 4, Case 2). The third case was characterised by an indicator of nitrate concentration <0 and an indicator of phosphate concentration >0 (Fig. 4, Case 3). The last case was characterised by an indicator of nitrate concentration <0 and an indicator of phosphate concentration <0 (Fig. 4, Case 4).

For each station, year-to-year changes in human nutrient over-enrichment were examined by contour diagrams with months as a function of years. The distance $d$ varied between 0 and 1. A value close to 0 indicated low disturbance whereas a value close to 1 reflected high nutrient over-enrichment (i.e. a high departure from the relative reference state, therefore the offshore site of a given station). The different cases were investigated for the three stations (Fig. 5). Then, we proposed a new indicator of human nutrient over-enrichment that was the aggregation of Cases 1, 2 and 3 (Fig. 6).

## 4. Results

### *4.1. Analyses 1: indicators of anthropogenic nutrient concentration*

Multiple Linear Regression (MLR) was used to remove the potential influence of climate on nutrient concentrations. Partial correlation coefficients were calculated to evaluate the influence of each variable (SST and silicates; keeping one of the explanatory variables constant to its mean) on nitrate and phosphate concentrations. All partial correlation coefficients calculated as part of the MLRs were significant with the exception of the correlation between phosphates and SST (keeping silicates constant) in Villefranche-sur-Mer ($r = -0.063$, $p = 0.287$; Table 2). Nitrate concentrations showed the strongest correlations with the two explanatory variables in all cases. For the three stations, the coefficients obtained with silicate concentrations were positive (from $r = 0.325$ to $r = 0.840$) whereas they were negative and generally weaker with SST (from $r = -0.063$ to $r = -0.748$). The coefficient of multiple determination was significant for all multiregressive models (Table 2). The addition of the second explanatory variable improved significantly the model in almost all cases ($p < 0.001$) with the exception of phosphate concentrations in Villefranche-sur-Mer where SST was added to the model after silicate concentration ($p$ of $F$-test = 0.322).

The examination of the coefficient of determination (Table 2) showed that the linear models explained well the variations in both nitrate and phosphate concentrations at both inshore and offshore sites in Wimereux (Fig. 2a and b). Because SST and silicate are highly determined by climate variability, the residuals of the multiregressive models enabled us to remove the influence of climate and to propose indicators of both nitrate and phosphate anthropogenic concentrations (Fig. 2c–f). Positive residuals should therefore be interpreted as a result of increasing nutrient concentrations from continent due to anthropogenic over-enrichment. The same analysis was conducted in Roscoff and Villefranche-sur-Mer (see Supplementary Figs. 1 and 2).

### *4.2. Analyses 2: mapping of the system state and both spatial and temporal comparisons*

The procedure enabled mapping of the relative reference state of the three monitored stations on which years were superimposed (Fig. 3 and Supplementary Figs. 3 and 4). While the relative reference states were larger from January to April in Wimereux and Roscoff (Fig. 3 and Supplementary Fig. 3) and from December to March in Villefranche-sur-Mer (Supplementary Fig. 4), they contracted the following months. On a year-to-year basis, the probability of each observation to belong to the relative reference state increased through time in Wimereux and Roscoff. A period of lower probability occurred until 2002 in Roscoff and 2003 in Wimereux, followed by an increase onwards. This result suggested a reduction in the anthropogenic over-enrichment from 2003 onwards in both sites.

In Villefranche-sur-Mer, probabilities also increased in winter. Years reported on the scatter plots were closer to the relative reference state except for a few observations (e.g. March 1997 and 1998, May to October 2008; Supplementary Fig. 4).

### *4.3. Analyses 3: year-to-year changes in human nutrient over-enrichment*

The 2-D Euclidean space was divided into four squares (we call cases hereafter, see Methods) to separate the respective influence of nitrates and phosphates on the calculation of the probability ($d = 1 - p$; see methods) of an observation to belong to the relative reference state. We present an example for Wimereux in January (Fig. 4; see the contour diagram performed for January in Fig. 3).

Case 1 (Fig. 4) presents the situation where the value of anthropogenic nitrate concentration was strictly superior to 0 (i.e. more concentration in nitrate inshore than offshore) and the value of anthropogenic phosphate concentration was strictly inferior to 0 (i.e. more concentration in phosphate offshore than

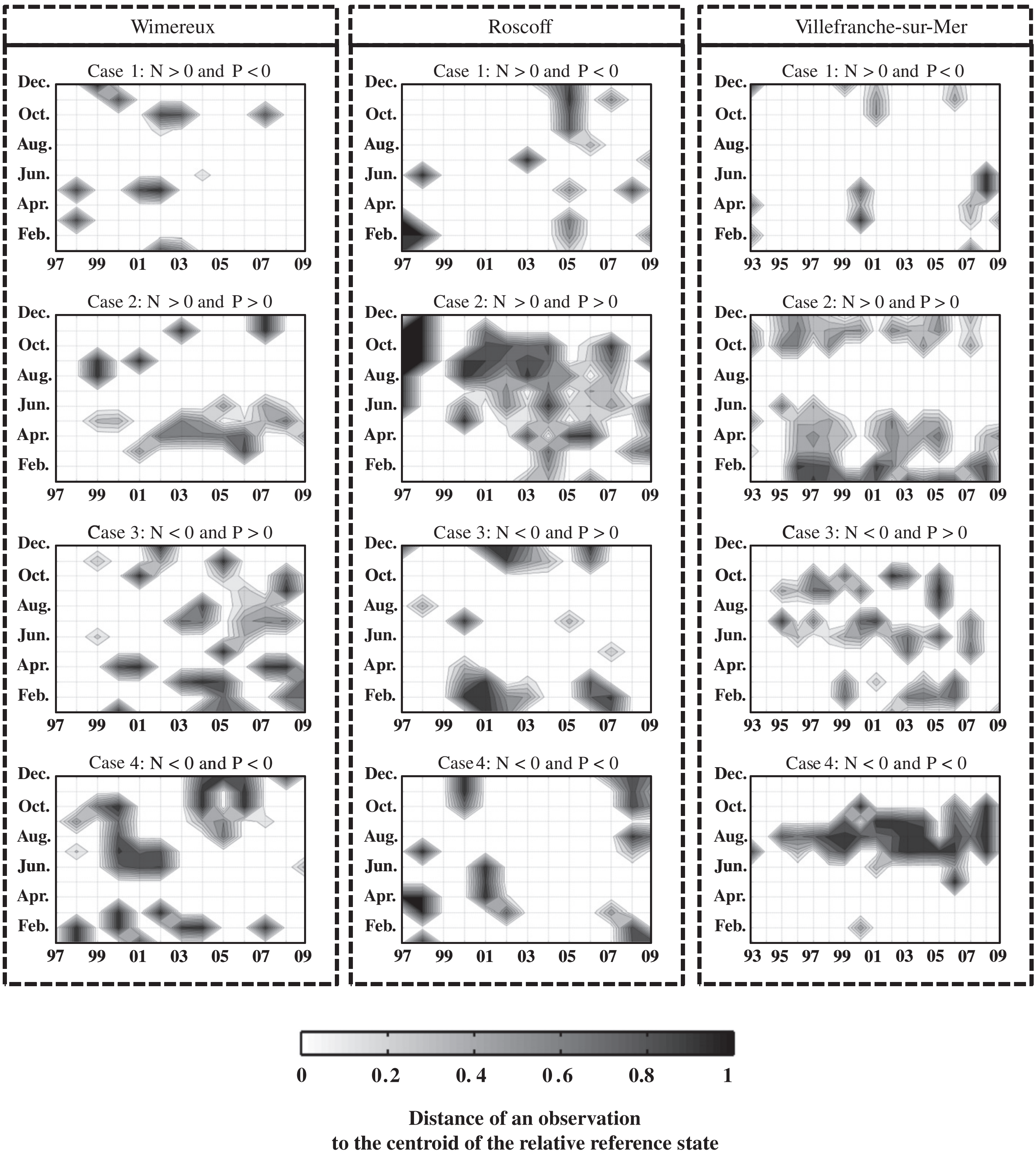


**Fig. 5.** Contour diagrams showing the temporal changes in the distance of an observation to the centroid of the relative reference states ($d = 1 - p$; see methods). Mapping of the distance was represented for the 4 cases (see Fig. 4) as a function of years, for the three studied sites from 1997 to 2009 for Wimereux (left panels) and Roscoff (middle panels) and from 1993 to 2009 for Villefranche-sur-Mer (right panels). The level of grey represents the distance of an observation to the centroid of the relative reference state. See Fig. 4 for the description of the different cases.

inshore). Although we could observe some episodic events, no clear pattern was observed in the three sites (Fig. 5; Case 1).

Case 2 (Fig. 4) presents the situation where both values of anthropogenic nitrate and phosphate concentrations were strictly superior to 0 (i.e. more concentration inshore than offshore). In Wimereux, an increase in nutrient was observed in spring from 2001 (Fig. 5 Case 2). In Roscoff, the increase in nutrient concerned all months from March to October. The intensity of the positive anomalies in nutrients decreased after 2004 (Fig. 5 Case 2). In Villefranche-sur-Mer, the increase in nutrient concentration

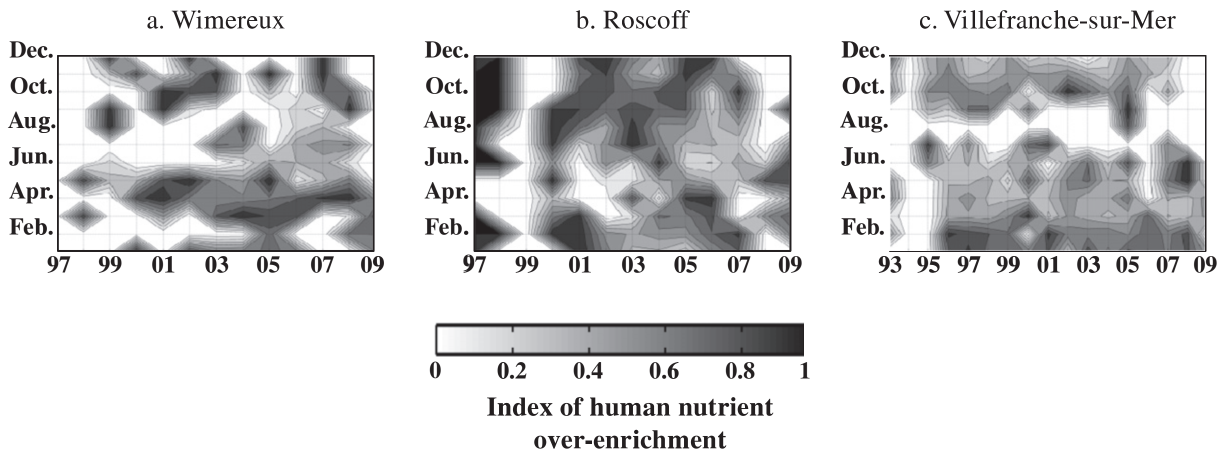


**Fig. 6.** Mapping of the index of human nutrient over-enrichment for the three studied sites as a function of months and years. (a) Wimereux from 1997 to 2009; (b) Roscoff from 1997 to 2009; (c) Villefranche-sur-Mer from 1993 to 2009. The indices of human nutrient over-enrichment corresponded to situations for which concentrations in anthropogenic nitrate and/or phosphate were superior to 0.

**Table 2**
Results from partial linear correlations and multiple linear regressions.

| | | Partial correlations | | | | Multi-regression | | |
|---|---|---|---|---|---|---|---|---|
| | | Temperature | | Silicate concentrations | | Temperature & silicate concentrations | | |
| | | *r* | *p* | *r* | *p* | *R* | *p* | *df* |
| Wimereux | Nitrate concentrations | −0.748 | <0.001 | 0.726 | <0.001 | 0.861 | <0.001 | 208 |
| | Phosphate concentrations | −0.492 | <0.001 | 0.667 | <0.001 | 0.755 | <0.001 | 208 |
| Roscoff | Nitrate concentrations | −0.759 | <0.001 | 0.840 | <0.001 | 0.942 | <0.001 | 233 |
| | Phosphate concentrations | −0.226 | <0.001 | 0.636 | <0.001 | 0.751 | <0.001 | 233 |
| Villefranche-sur-Mer | Nitrate concentrations | −0.418 | <0.001 | 0.390 | <0.001 | 0.586 | <0.001 | 287 |
| | Phosphate concentrations | −0.063 | 0.287 | 0.325 | <0.001 | 0.353 | <0.001 | 287 |

*r*: values of correlations, *p*: probabilities, *R*: coefficients of multiple correlation, *df*: degrees of freedom.

remained driven by the seasonal climatic variability with maximal nutrient concentrations occurring during winter and spring (Fig. 5 Case 2).

Case 3 (Fig. 4) presents the situation where the value of anthropogenic nitrate concentration was strictly inferior to 0 (i.e. more concentration in nitrate offshore than inshore) and the value of anthropogenic phosphate concentration strictly superior to 0 (i.e. more concentration in phosphate inshore than offshore). Contrary to Case 1, the frequency of such events was more widespread in all sites (Fig. 5 Case 3). In Wimereux, an increase in phosphate concentration associated to a reduction of nitrate was frequently observed after 2003. In Roscoff, some episodic events occurred mainly in winter between 2000 and 2007 (Fig. 5 Case 3). In Villefranche-sur-Mer, these events were mainly detected in February (between 2003 and 2007), June and September (Fig. 5 Case 3).

Case 4 (Fig. 4) presents a perhaps unexpected situation where indicators in both nitrate and phosphate anthropogenic concentrations were strictly inferior to 0 (i.e. more concentration in both nitrate and phosphate offshore than inshore; Fig. 4). A strong value in the distance reflected here a strong negative anomaly in inshore nutrient concentration. The inshore site in Villefranche-sur-Mer displayed a pronounced seasonal pattern characterised by a deficit of both nutrients from June to October (Fig. 5; Case 4). The examination of Atlantic inshore sites revealed some depletions more difficult to interpret. In Wimereux (Fig. 5; Case 4), year-to-year fluctuations in negative inshore anomalies were more pronounced in summer from 2000 to 2003, in autumn between 2004 and 2007 and to a lesser extent in winter for some years (e.g. 1998, 2000; see Fig. 5; Case 4). No clear pattern was observed in Roscoff (Fig. 5; Case 4).

An indicator was then designed, resulting from the combination of Cases 1, 2 and 3, to provide a measure of human nutrient over-enrichment in inshore coastal systems (Fig. 6). In Wimereux (Fig. 6a), we observed a clear increase in nutrient concentrations caused by both nitrate and phosphate whereas the increasing anomalies for the rest of the period were mainly the result of an increase in phosphate inputs since 2003 (see Fig. 5; Case 3). In Roscoff, human nutrient over-enrichment was more widespread and mainly detected from summer to winter (Fig. 6b). A decrease in the anomalies in nitrate and phosphates inshore concentrations was detected after 2004. As seen earlier, the indicator of human nutrient over-enrichment was more driven by seasonal climatic variability in Villefranche-sur-Mer (Fig. 6c).

## 5. Discussion and conclusions

It is now widely accepted that the last 200 years have been periods of particularly rapid and pronounced environmental changes (e.g. Omori et al., 1994; Kremer et al., 2005). Among all systems of the biosphere, coastal systems are one of the most valuable and vulnerable of Earth's habitats (Jickells, 1998). Historically, developing human civilisations has often been concentrated in coastal areas where access to water promoted trade, commerce, or use of wastes (e.g. Van Andel, 1981; Vitousek et al., 1997b). This tendency has significantly increased through time (Small and Nicholls, 2003). At a global scale, both coastal and estuarine waters have thereby been affected by anthropogenic activities causing strong alterations of some natural biogeochemical cycles (Cloern, 2001; Borja and Dauer, 2008). Comparing a hypothetical undisturbed system (as that defined by Billen and Garnier, 2007) to various present conditions, disproportionate nutrient loadings into coastal waters have been noticed with a 6–50 times increase for nitrate loading and a 18–180 times increase for phosphate loading for systems such as the Baltic Sea, the Chesapeake Bay and the Eden Bight (Conley, 2000). These human perturbations have resulted in detrimental effects on the health of coastal systems (e.g. Sherman and Duda, 1999; Smith et al., 2003). For example, eutrophication and its related consequences (e.g. hypoxia and anoxia) are considered as major issues in coastal seas (Diaz, 2001; Gray et al., 2002; Selman et al., 2008).

Not a single coastal system remains unaffected by human activities (Richardson and Poloczanska, 2008) and it is still challenging to tease apart the influence of anthropogenic forcing to that related to climate variability (Alley et al., 2003). Although modern day nutrient budgets are available for some estuarine and coastal systems (e.g. Nixon, 1995), few estimates are available to compare present day nutrient loading to those under undisturbed conditions (Conley, 2000). Furthermore, few classical statistical techniques are able to identify, detect and quantify abrupt or progressive modifications in nutrient concentrations. Most attempts to quantify the impact of human activities on nature have focused on the development of ecological indicators (García et al., 2010). Some methods have also been designed to evaluate the status of coastal waters focussing on different issues such as impacts on human health and/or ecosystem function (e.g. Luck et al., 2003; Borja and Dauer, 2008). While most of the algorithms cannot cope with missing values and associated biases, the technique presented in this study is insensitive to missing data and is unlikely to be influenced significantly by temporal autocorrelation (Goberville et al., 2011). This technique allows the building of relative reference states, which in turn enable each observation to be compared. While our procedure does not require long-term data to design the relative reference state, long-term data enable a better quantification of the noise inherent to a sampling site and thereby increase the signal to noise ratio, allowing a potential perturbation to be better detected. The resulting probability directly quantifies the intensity of the perturbation and provides indicators of human nutrient over-enrichment on coastal systems. There is no limitation on the number of variables although a too high number may increase the risk of multicollinearity when the Mahalanobis distance is calculated and the procedure can be virtually coupled on all monitoring programmes and easily be transposed to any data sets, parameters or pollutants (e.g. heavy metals, hydrocarbons). Furthermore, our procedure can be applied in all types of systems, be equatorial or polar. In a tropical system, the variance around the mean is likely to be smaller than that of an extratropical system.

Through the proposed indicators of human nutrient over-enrichment, our results present a new way to quantify a perturbation in natural systems. After disentangling the influence of natural variability on nutrients, residuals in both nitrate and phosphate concentrations were used to monitor the potential influence of human activities on nutrient concentration. However, two limitations must be considered before interpreting our results. First, both offshore and inshore sites have probably been altered by anthropogenic activities. The anthropogenic influence on surface waters has greatly affected global biogeochemical cycles, increasing natural nutrient levels even offshore (Meybeck, 1982; Vitousek et al., 1997a). In this study, we assumed that the offshore site can be considered as a relative reference state, even if it has probably been impacted by human nutrient over-enrichment. The implication is that our quantification of human nutrient over-enrichment is probably conservative. Second, as some recent studies did (e.g. Cloern, 2001; Billen and Garnier, 2007; Ludwig et al., 2009), we chose to focus exclusively on both nitrate and phosphate anthropogenic concentrations to evaluate the human nutrient over-enrichment and to detect potential alteration of the current states of French coastal systems for the period 1997–2009. Despite the use of a limited number of parameters (a total of 13 parameters are currently available in SOMLIT), a consistent anthropogenic signal emerged from our indicators of human nutrient over-enrichment.

The three stations we studied showed two distinctive seasonal periods in nitrate, phosphate and silicate concentrations. During winter, nutrients behave as conservative elements. In contrast during summer, they are consumed by phytoplankton in the surface layer (Rippeth and Jones, 1997). Nutrient concentrations were not similar in all stations. Nitrates were between 3 and 6 times more concentrated and phosphates between 2 and 4 times superior in the sites of the English Chanel than in the Mediterranean site. In the eutrophic areas, phytoplankton growth might be light-limited amplifying the conservative behaviour in nutrients in winter (Walne, 1993). This is corroborated by the high partial correlations between nitrates or phosphates and temperature or silicates (Table 2, Fig. 2 and Supplementary Fig. 1). Phytoplankton was more nutrient-limited in the oligotrophic site, temperature limitation not being relevant as the temperature ranges from 12 °C to 28 °C are sufficient for primary production (Guizien et al., 2007). In this site, surface waters are depleted in phosphates, nitrates and silicates due to anti-estuarine water circulation (Béthoux et al., 2002), scarcity in freshwater inputs and the influence of Coriolis force on the Liguro-provencal current circulation (Font et al., 1988). Furthermore, as shown by the weak partial correlations in Villefranche-sur-Mer, dissolved nutrients are not directly transported in flood waters and can be released at coastal waters through reactive processes of suspended matters (Guizien et al., 2007). Conversely, the two eutrophic sites are clearly impacted by inlets in freshwater from the closest rivers (see Table 1) and intensive oceanic inflow from the North Atlantic (e.g. 82.68% of nitrogen input in the English Channel originated from this phenomenon; Jickells, 1998).

The removal of the climatic influence on the oligotrophic site was less efficient than for eutrophic sites because we found a clear seasonal variability in the indicator of human nutrient over-enrichment for this region (Fig. 6c). The persistent seasonal variability suggests that human nutrient over-enrichment in this Mediterranean site was not predominantly modulated by temperature and continental influence. Furthermore, peculiar mechanisms such as precipitation of phosphate with iron containing dust from Sahara (Krom et al., 1991) and nitrogen fixation (Béthoux et al., 1998) make it more complex the biogeochemistry in this area. An excess in phosphate anthropogenic concentration occurred during algal blooms (April–June and September–October; Fig. 5; Case 3), a result in agreement with the strong limitations in nitrate already observed in the upper photic zone (Krom et al., 1991; Thingstad et al., 1998).

In contrast to the Mediterranean site, no climatic influence remained from the indicators of human nutrient over-enrichment in the sites of the English Channel (Fig. 2 and Supplementary Fig. 1). External loadings from the adjacent open sea and from continents are a major component of the budget in nitrates and phosphates (Jickells, 1998; De Galan et al., 2004; Halpern et al., 2008; Vermaat et al., 2008). In Roscoff, the indicators of human nutrient over-enrichment exhibited recurrent nutrient loadings (Fig. 6b) whereas in Wimereux, they occurred mainly from March to June (Fig. 6a). The variation in human nutrient over-enrichment in both sites could be explained by local peculiarities. By its runoff near the inshore site in Roscoff, the Penzé River provides high loading of nutrients in estuaries because of the intense use of fertilisers by agriculture in Brittany (Desalos, 1999). In this area, strong and constant loadings have occurred throughout the year (generally greater than 400 $\mu mol\ L^{-1}$ for nitrates even in summer; Wafar, 1981). Our results enable the excessive proliferations of species such as Ulva sp. in some coastal ecosystems to be explained. For example, these proliferations have been detected during spring and summer near the mouth of the Penzé River (Dussauze and Ménesguen, 2008). In Wimereux, the human nutrient over-enrichment has rather occurred predominantly in spring, during the period of agricultural activity.

In many countries, legislation mandates assessment of the water chemistry, biota, and physical environment of ecosystems, many of which have been highly impacted by human activities (Gergel et al., 2002). Severe actions have been undertaken to reduce the nutrient loadings from rivers (e.g. the Clean Water Act of the United States, the Water framework Directive of the European Union) but such actions have not always been successful. Despite the many conferences on the issue, such as the 4th North Sea Conference Progress Report (1995) where management plans were elaborated, nitrate and phosphate concentrations in the water of the two stations in the English Channel did not exhibit significant decreasing trends between 1997 and 2009. Similar results have also been observed along the Belgian coast (De Galan et al., 2004).

Past efforts to identify changes in marine coastal systems used either coarse categorical or perhaps too specific numerical tools (Ban and Alder, 2008; Halpern et al., 2008). The library of empirical studies that relate anthropogenic impacts on coastal systems is increasing. The development of methods and indicators is crucial to detect more rapidly potential human-induced alterations and to quantify their intensity. Considering this issue, the method and the indicators of human nutrient over-enrichment proposed in this study attempt to deal with these actual scientific requirements. These new indicators enabled the quantification of the anthropogenic influence on nutrient concentrations and allow us to test immediately an observation by the knowledge of a relative reference state determined empirically or alternatively fixed by expertise. Developing statistical procedures to detect perturbations as soon as data become available, implementation of monitoring network and relevant indicators of human nutrient over-enrichment all appear to be important for more appropriate management strategies in the future (Niemeijer, 2002; Pereira and Cooper, 2006; Richardson and Poloczanska, 2008).

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## Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.marpolbul.2011.05.024.

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