# Estimation of the Human Impact on Nutrient Loads Carried by the Elbe River

Mohamed Hussian, Anders Grimvall and Wilhelm Petersen

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<sup>1</sup>Mohamed Hussian, <sup>1</sup>Anders Grimvall and <sup>2</sup>Wilhelm Petersen <sup>1</sup>Department of Mathematics, Linköping University

<sup>2</sup>Institute for Coastal Research, GKSS Research Center Germany

Address for correspondence: Department of Mathematics, Linköping University, SE-58183 Linköping, Sweden; mohus@mai.liu.se.

#### Abstract

The reunification of Germany led to dramatically reduced emissions of nitrogen (N) and phosphorus (P) to the environment. The aim of the present study was to examine how these exceptional decreases influenced the amounts of nutrients carried by the Elbe River to the North Sea. In particular, we attempted to extract anthropogenic signals from time series of riverine loads of nitrogen and phosphorus by developing a normalization technique that enabled removal of natural fluctuations caused by several weatherdependent variables. This analysis revealed several notable downward trends. The normalized loads of total-N and NO<sub>3</sub>-N exhibited an almost linear trend, even though the nitrogen surplus in agriculture dropped dramatically in 1990 and then slowly increased. Furthermore, the decrease in total-P loads was found to be considerably smaller close to the mouth of the river than further upstream. Studying the predictive ability of different normalization models showed the following: (i) nutrient loads were influenced primarily by water discharge; (ii) models taking into account water temperature, load of suspended particulate matter, and salinity were superior for some combinations of sampling sites and nutrient species; semiparametric normalization models were almost invariably better than ordinary regression models.

# Contents

1	Intro	Introduction				
2	Study area and data sets					
3 Changes of the environmental pressure on the Elbe						
4	Statistical procedures					
	4.1	Quality assessment of collected data	6			
	4.2	Estimation of monthly and annual nutrient loads	6			
	4.3	Calculation of temerature and salinity indices	8			
	4.4	Normalization of nutrient loads	8			
	4.5	Selection of roughness penalty factors and assessment of				
		predictive ability of the tested models	9			
5	Results					
	5.1	Annual riverine loads of nutrients	10			
	5.2	Impact of factors other than water discharge on riverine loads	11			
		of nutrients				
	5.3	The predictivity of different normalization models	12			
	5.4	Normalized loads of nitrogen	13			
	5.5	Normalized loads of phosphorus	17			
6	Discussion and conclusions					
	6.1	Methodological aspects of the normalization of riverine loads	19			
		of nutrients				
	6.2	Extracted anthropogenic signals	20			
7	Ackı	Acknowledgment				
8	Literature					

# 1 Introduction

The reunification of Germany in 1990 triggered dramatic changes in the flows of nutrients through the Elbe River Basin (Behrendt, 1999; ARGE, 2001). The direct emissions of phosphorus to water decreased to a fraction of their previous levels, and there was also a significant decrease in the point emissions of nitrogen. Changes in the agricultural practices were equally remarkable, which, for example, caused the nitrogen surplus in the top soil to drop by more than 50% in the German part of the Elbe Basin. Hence, it is not surprising that the riverine loads of nutrients were considerably lower in 1993–1997 than in 1983–1987. Nevertheless, it has not yet been clarified how rapidly the Elbe River responded to these exceptional changes in human activities in the drainage area, or how the response varied with the sampling site and species of nitrogen or phosphorus under consideration. Moreover, it has not been determined what type of statistical data analysis can most efficiently distinguish between natural fluctuations and the effects of human interventions.

Over the past few years, scientists have found it of increasing interest to normalize environmental data, that is, to remove from collected data any variation that can be attributed to natural fluctuations. A theoretical framework for such adjustment has recently been established (Grimvall *et al.*, 2001). In addition, a number of research articles have been devoted to normalization of different types of environmental data, and most of them have dealt with meteorological normalization of air quality variables, in particular the concentration of tropospheric ozone (Bloomfield *et al.*, 1996; Shively & Sager, 1999; Gardner & Dorling, 2000*a*,*b*; Thompson *et al.*, 2001). However, flow normalization of riverine loads of nutrients has also been addressed (Stålnacke *et al.*, 1999; Stålnacke & Grimvall, 2001; Uhlig & Kuhbier, 2001), and OSPAR (the Oslo-Paris Commission) has established a working group for hydrological normalization of riverine inputs to the North Sea. The objectives of the present study were as follows:

- to demonstrate how information on water discharge, water temperature, load of suspended particulate matter (SPM), and salinity can be used to normalize riverine loads of different species of nitrogen and phosphorus;
- to compute the load of nutrients carried by the Elbe to the North Sea and decompose the observed interannual variation into natural fluctuations and effects triggered by the reunification of Germany;
- (iii) to compare the anthropogenic signals that can be extracted from time series of riverine loads of different nitrogen and phosphorus species at different sampling sites in the lower reaches of the Elbe.

In particular, we modified and evaluated the semiparametric normalization technique developed by Stålnacke and Grimvall (2001). Furthermore, we critically examined whether the goal of a 50% reduction of nutrient loads has been achieved.

## 2 Study area and data sets

The Elbe River Basin has a total area of 148,268 km<sup>2</sup> and includes the cities of Prague, Dresden, Berlin, and Hamburg (Figure 1). The present study focused on temporal changes in nutrient loads carried by the Elbe at one site (Schnackenburg) upstream of Hamburg and three sites (Grauerort, Brunsbüttel, and Cuxhaven) downstream of that city (Figure 2). More precisely, monthly nutrient loads were computed by combining nutrient concentrations measured at these sites with daily water discharge data recorded at NeuDarchau, 52 km downstream of Schnackenburg. Table I shows the number of observations and the time span of the water quality measurements at the different sampling sites. All of the analyzed data were obtained from 'Arbeitsgemeinschaft Elbe' (ARGE) in Hamburg.



Figure 1. The Elbe River Basin.



Figure 2. Sampling sites for water quality measurements in the lower reaches of the Elbe.

Sampling site	Time span	Number of observations
Schnackenburg	1988–2000	422–436
Grauerort	1988–2000	386–424
Brunsbüttel	1985–2000	546–549
Cuxhaven	1992–2000	222–229

**Table I**. Time span and number of water quality observations (nutrients, chloride, and suspended particulate matter).

# 3 Changes in the environmental pressure on the Elbe

The nitrogen surplus is the difference between the amount of nitrogen added to the top soil and the amount of that element removed through harvesting, and this value is often used as an indicator of the potential loss of nitrogen from soil to water (Wendland *et al.*, 1998; 2001). Figure 3 illustrates how dramatically this indicator decreased from 1989 to 1990 in the German part of the Elbe Basin. During the past few years, an increase has been noted, but the present level is comparable to the nitrogen surplus recorded in the 1960s. The changes in point emissions of phosphorus and nitrogen are shown in Table II.



**Figure 3**. Average annual nitrogen surplus on agricultural land in the German part of the Elbe River Basin (ARGE, 2001).

	1983-			
Point emissions (kton/yr)	Ν	Р	Ν	Р
Direct industrial discharge	59.8	2.8	18.0	0.5
Discharge from wastewater treatment plants	69.0	12.5	45.6	4.4

 Table II. Point emissions of nitrogen (N) and phosphorus (P) to the Elbe River (ARGE, 2001)

# 4 Statistical procedures

#### 4.1 Quality assessment of collected data

All nutrient concentration data were inspected by plotting observed values against time (decimal year), day of the year (Julian day), water discharge, and suspended particulate matter (SPM). Furthermore, total nitrogen and phosphorus values were plotted against values of the inorganic fraction of these elements, and the values recorded at the four sampling sites were compared with each other. In a few cases, the sum of the concentrations of the inorganic nitrogen species exceeded the measured amounts of total nitrogen. In addition, a small number of outliers was detected. Thus a special study was conducted to determine whether this in any way influenced the final results of the data analysis (see below).

#### 4.2 Estimation of monthly and annual nutrient loads

Monthly and annual nutrient loads were calculated by first expanding the time series of observed concentration data to complete series of daily data and then summing daily values of the product of concentration and water discharge. The concentration data were expanded using a nonparametric smoothing procedure (LOESS; Cleveland & Devlin, 1988). When the smoothing parameter (or bandwidth) of this procedure was set to a minimum, the expanded values were identical to those obtained when concentration values were connected with straight lines. Higher values of the smoothing parameter implied that outliers were suppressed (Figure 4). However, as long as the smoothing did not attenuate the seasonal pattern in observed concentration data, the choice of smoothing parameter had only a minor impact on the computed monthly and annual nutrient loads (Figure 5).



**Figure 4**. Observed and smoothed daily values of the concentration of total nitrogen at Grauerort. The two smoothed curves respectively represent bandwidths of 0.01 and 0.05.



**Figure 5**. Estimated monthly loads of total nitrogen at Grauerort obtained using two different smoothing parameters (bandwidths) to expand measured concentration data to a complete series of daily data.

#### 4.3 Calculation of temperature and salinity indices

It is well known that NH<sub>4</sub>-N present in wastewater is readily transformed into NO<sub>x</sub> species under consumption of oxygen (Wolter *et al.*, 1985), and the velocity of this

nitrification process is strongly influenced by water temperature. Moreover,  $NH_4$  is the nitrogen species that is first consumed by algae. We found that after 1991, when oxygen was normally in good supply, the observed concentrations of  $NH_4$ -N decreased almost exponentially with the water temperature (Figure 7). Accordingly, we computed a monthly temperature index of the form

$$T_{\mu} = \sum_{k} \exp(-\mu T_{k}),$$

where  $T_k$  depicts daily temperature values, and  $\mu$  is estimated by fitting an exponential function to observed pairs of water temperature and concentration values. The summation in the formula ranges over all days of the month for which the index is computed.

The samples collected at Cuxhaven were often strongly affected by sea water, and that phenomenon was also occasionally noted at Brunsbüttel and Grauerort. Therefore, flow-weighted chloride concentrations were computed for each month and were used as indicators of a marine influence.

#### 4.4 Normalization of nutrient loads

Monthly values of nutrient loads were normalized by employing a semiparametric regression model to remove the temporal variation that could be attributed to fluctuations in water discharge, load of SPM, water temperature, and salinity. Our model had the general form

$$y_{ij} = \alpha_{ij} + \beta_{1,j} x_{1,ij} + \dots + \beta_{p,j} x_{p,ij} + \varepsilon_{ij}, \quad i = 1, \dots, n \quad j = 1, \dots, m$$

where  $y_{ij}$  is the observed response (nutrient load) for the *j*th month of the *i*th year,  $x_{k,ij}$ ,  $k=1, \ldots, p$  represent contemporaneous values of p explanatory variables standardised to mean zero and variance one, and  $\varepsilon_{ij}$  is a random error term with mean zero. The slope parameters ( $\beta_{k,j}$ ,  $k=1, \ldots, p$ ) were permitted to vary with the season (*j*) under consideration, and the intercept ( $\alpha_{ij}$ ) was permitted to vary with both season (*j*) and year (*i*). However, rapid changes in the intercept were controlled by so-called roughness penalty factors ( $\lambda_1$  and  $\lambda_2$ ), and the intercept and slope parameters were estimated by minimizing the expression

$$\begin{split} S(\alpha,\beta) &= \sum_{i,j} (y_{ij} - \alpha_{ij} - \beta_{1,j} x_{1,ij} - \dots - \beta_{p,j} x_{p,ij})^2 + \lambda_1 \sum_{i,j} (\alpha_{ij} - \frac{\alpha_{i+1,j} + \alpha_{i-1,j}}{2})^2 \\ &+ \lambda_2 \sum_{i,j} (\alpha_{ij} - \frac{\alpha_{i,j-1} + \alpha_{i,j+1}}{2})^2, \end{split}$$

where the first sum ranges over all values of i and j for which both the response variable and the explanatory variables have been observed. A univariate form of this model was first used by Stålnacke and coworkers (1999), and detailed information about algorithms for parameter estimation has been published by Stålnacke and Grimvall (2001).

# 4.5 Selection of roughness penalty factors and assessment of the predictive ability of the tested models

The penalty factors  $\lambda_1$  and  $\lambda_2$  were determined by cross-validation. With this technique, the entire data set is separated into an estimation set (or training set) and a test set. The model is first fitted to the estimation set and is subsequently used to predict the observations in the test set, that is, the values that have been left out of the estimation step. We defined *m* estimation sets  $M_i$ , *i*=1, ..., *m* by leaving out one-year-long blocks of observations, and then we computed a so-called PRESS-value (i.e., a sum of squared prediction errors):

$$S(\lambda_1, \lambda_2) = \sum_{i} \sum_{(i,j) \notin M_i} (y_{ij} - \hat{\alpha}_{ij} - \hat{\beta}_{1,j} x_{1,ij} - \dots - \hat{\beta}_{p,j} x_{p,ij})^2.$$

Finally, the factors  $\lambda_1$  and  $\lambda_2$  were selected in such a way that  $S(\lambda_1, \lambda_2)$  was minimized, and the corresponding Root Mean *PRESS* value

$$min\left\{\sqrt{\frac{1}{N}S(\lambda_1,\lambda_2)};\lambda_1>0,\lambda_2>0\right\}$$

was used as a measure of the predictive ability of the normalization model under consideration.

# **5** Results

### 5.1 Annual riverine loads of nutrients

Figure 6 shows estimated annual loads of nutrients for different sampling sites and species of nitrogen and phosphorus. A downward tendency in nutrient loads can be discerned in most of the time series of data. However, the interannual variation is large, and the annual loads vary strongly with the annual water discharge values.



**Figure 6**. Estimated annual loads of nitrogen and phosphorus for different sampling sites shown together with water discharge values from NeuDarchau.

# 5.2 Impact of factors other than water discharge on riverine loads of nutrients

Figure 7 shows how the concentration of NH<sub>4</sub>-N varied with water temperature at Schnackenburg, and, in particular, it can be seen that the concentration of this nitrogen species was very low at temperatures exceeding 10 °C. The concentration of total phosphorus was correlated to the concentration of SPM, which can be explained by the fact that a substantial fraction of the phosphorus found in surface waters is bound to particles. Considering the scatter chart in Figure 8, the highest concentrations of phosphorus at Brunsbüttel occurred in samples with a high level of SPM.



**Figure 7**. Scatter chart of daily concentrations of  $NH_4$ -N plotted against water temperature at Schnackenburg, 1992 - 2000. The fitted curve has the equation y=1.1341\*exp(-0.1869\*x).



**Figure 8**. Scatter chart of daily concentrations of total phosphorus and suspended particulate matter (SPM) at Brunsbüttel, 1985–2000.

### 5.3 The predictive ability of different normalization models

Using simple linear regression, it was easy to show that a substantial fraction of the temporal variation in monthly loads of nitrogen and phosphorus represented natural fluctuations in water discharge. Closer examination of the predictive ability of different parametric and semiparametric normalization models indicated that, for some combinations of nutrient species and sampling sites, the load was also related to water temperature, salinity, or amount of SPM (see Table III). As expected, we found that information about SPM was when normalizing the load of total phosphorus, and that nutrient loads recorded at sites in the mixing zone of river and sea water were influenced by the salinity. The impact of water temperature on the load of NH<sub>4</sub>-N was not revealed until the analysis was restricted to the period 1992–2000; before that time, the influence of temperature was overshadowed by other factors controling the growth of algae.

**Table III**. The predictive ability of different parametric and semiparametric normalization models. The Root Mean *PRESS* values in the table refer to the roughness penalty factors  $\lambda_1$  and  $\lambda_2$ , which produced the smallest sum of squared prediction errors.

	Sampling site	Ordinary	Semiparametric	Optimal semiparametric	
		regression	regression on	regression model	
Nutrient		on discharge	discharge	Evolanatory	Poot Mean
		Root Mean	Root Mean		
		PRESS	PRESS	variables	FRESS
	Schnackenb.	0.333	0.161	Discharge, SPM	0.155
Total N	Grauerort	0.288	0.170	Discharge	0.170
TOTAI-IN	Brunsbüttel	0.285	0.161	Discharge, SPM	0.152
	Cuxhaven	0.278	0.255	Discharge	0.255
	Schnackenb.	0.239	0.217	Discharge	0.217
	Grauerort	0.259	0.155	Discharge	0.155
NO3-IN	Brunsbüttel	0.276	0.217	Discharge, SPM, Cl	0.205
	Cuxhaven	0.307	0.219	Discharge, Cl	0.200
	Schnackenb.	1.096	0.407	SPM	0.397
	Grauerort	1.007	0.422	Discharge	0.422
INF14-IN	Brunsbüttel	0.971	0.571	Discharge	0.571
	Cuxhaven	0.804	0.700	Discharge, Cl	0.484
	Schnackenb.	0.738	0.547	Discharge, SPM	0.490
Tot D	Grauerort	0.659	0.627	Discharge, SPM	0.299
TUI-F	Brunsbüttel	0.627	0.574	Discharge, SPM	0.570
	Cuxhaven	0.418	0.487	Discharge, Cl	0.473
	Schnackenb.	1.084	0.652	Discharge	0.652
	Grauerort	0.899	0.625	Discharge	0.625
PU4-P	Brunsbüttel	0.669	0.547	Discharge, SPM	0.403
	Cuxhaven	0.539	0.629	Discharge, SPM	0.627

## 5.4 Normalized loads of nitrogen

After normalization, monotone downward trends emerged in all time series of riverine loads that encompassed values from the late 1980s to 2000. Moreover, the normalization was successful in the sense that the interannual variation in the normalized loads was much smaller than in the time series of observed loads. In general, there was

also good agreement between the results obtained for different sampling sites, although some discrepancies were noted and further investigated.

Figure 9 shows that the normalized annual loads of total nitrogen and  $NO_3$ -N were almost identical for the single sampling site upstream of Hamburg (i.e., Schnackenburg) and the two sites immediately downstream of that city (i.e., Grauerort and Brunsbüttel). Also, it can be seen that there was a gradual decrease in total nitrogen from 1990 to 2000, despite the fact that the agricultural nitrogen surplus dropped dramatically in 1990 and then slowly increased. The somewhat different results for the third downstream site (Cuxhaven) can easily be explained by the marine influence at that location and biochemical reactions in the Elbe estuary.

Figure 10a indicates that the normalized loads of NH<sub>4</sub>-N first decreased markedly and then leveled out in the early 1990s. However, some features of these values required closer scrutinization. For example, some of the normalized values for Brunsbüttel appeared to be unrealistically low, and there was a peak at Grauerort in 1996. Further examination of different subsets of the collected data revealed that regression-based normalization can be problematic when the temporal changes are so dramatic that the relationships between nutrient loads and the explanatory variables change significantly over the study period. A new data analysis that was restricted to the period 1992–2000 produced more credible normalized loads (Figure 10b). Furthermore, the peak in 1996 vanished, because the new normalization model discerned a relationship between NH<sub>4</sub>-N loads and water temperature, and there was an unusually long period of low temperature values for that particular year.



**Figure 9**. Normalized annual loads of total nitrogen (a) and  $NO_3$ -N (b) at the four investigated sampling sites along the Elbe. The illustrated results were obtained using the optimal semiparametric models listed in Table III.



**Figure 10**. Normalized annual loads of NH<sub>4</sub>-N at the four investigated sampling sites along the Elbe River. a) Diagram showing the results obtained using the optimal models in Table III to normalize all available data. b) Diagram illustrating the results of analysis restricted to data from 1992–2000 and using water discharge, water temperature, and load of SPM as explanatory variables.

### 5.5 Normalized loads of phosphorus

Figure 11 shows how the normalized load of  $PO_4$ -P at Schnackenburg first declined dramatically from 1989 to 1991 and then continued to decrease at a slower rate.

By comparison, temporal trends downstream of Hamburg at Grauerort and Brunsbüttel differed in that the initial decrease was smaller, and it is difficult to discern any downward trend at all after 1993.



**Figure 11**. Normalized annual loads of PO<sub>4</sub>-P at the four investigated sampling sites along the Elbe River. The normalization was carried out using the optimal semiparametric models listed in Table III.

Also, the trend in total phosphorus was steeper at Schnackenburg upstream of Hamburg than at any of the downstream sampling sites (Figure 12). Furthermore, it can be noted that the load of total phosphorus was larger at Brunsbüttel than at the other three sites. Both these phenomena can be explained by sedimentation and resuspension of particulate matter. Compared to the other sites, Schnackenburg is closer to the areas where the major emission reductions took place around 1990. Brunsbüttel is situated in the mixing zone of river and sea water, and it is well known that tides can cause an upstream transport of marine particles (tidal pumping) that can inflate the loads recorded at such locations (Schwedhelm *et al.*, 1988).

Closer examination of the data revealed that the normalized annual load of total phosphorus at Brunsbüttel reached a minimum in 1993, a year with an unusually small number of high chloride values. Further analysis of collected data showed that the minimum in 1993 vanished when the normalization was performed using data restricted to 1992–2000 and salinity as an explanatory variable.



**Figure 12**. Normalized annual loads of total phosphorus at the four investigated sampling sites along the Elbe River. a) Diagram showing results obtained using the optimal models in Table III to normalize all available data. b) Diagram illustrating analysis restricted to data from 1992–2000 and using water discharge, salinity, and load of SPM as explanatory variables.

### 6 Discussion and conclusions

The purpose of normalizing time series of environmental data is to clarify anthropogenic impacts by removing natural fluctuations from the collected data. In the present study, relatively simple statistical normalization models were able to explain and remove almost all irregular year-to-year variation in the loads of nutrients carried by the Elbe. As expected, the loads of both nitrogen and phosphorus species were strongly influenced by water discharge. However, it was also apparent that variation in water temperature, salinity, and load of SPM could explain a substantial part of the total variation in riverine loads. Hence, there is a need for statistical procedures that can adjust measured data for natural fluctuations in several explanatory variables. The following discussion initially deals with methodological aspects of normalization and thereafter concerns the anthropogenic signals that were extracted from data on the Elbe.

# 6.1 Methodological aspects of the normalization of riverine loads of nutrients

An ideal normalization procedure should remove all natural fluctuations from measured data and leave the remaining variation unchanged. However, this goal is impossible to achieve for the following reasons:

- (i) normalization may suppress anthropogenic signals that coincide temporally with natural fluctuations;
- the variables used as indicators of natural fluctuations may include anthropogenic components.

To lessen the first problem, it is important to make the normalization models as simple as possible. We used cross-validation and Root Mean PRESS values to select normalization models. More precisely, we used test sets representing one-year-long time periods, because the so-called leave-one-out procedure may result in unnecessarily complex models when observed data are serially correlated (Shao, 1993; Libiseller & Grimvall, 2002). The severity of the second problem is related to the length of the study period. It is indisputable that human-induced changes in climate and land-use can cause long-term trends in water discharge. Nevertheless, it is reasonable to assume that the variation in water discharge in the Elbe from 1985 to 2000 was primarily of natural

origin. It also seems probable that, during the same period, the fluctuations in water temperature and salinity had only minor anthropogenic components. The variation in load of SPM may include more substantial effects of human activities. However, the short duration of the SPM peaks strongly indicates that most of the observed variation represented natural fluctuations caused by specific events of precipitation and mixing of freshwater and sea water. Accordingly, we conclude that all normalization models used in the present study can be justified.

Closer examination of the predictive ability of different normalization models clearly demonstrated that, in general, the semiparametric approach developed in this paper was superior to normalization based on ordinary least squares regression. However, two common features of regression-based normalization techniques require further discussion. First, normalization is based on the idea that there are relatively stable relationships between the response variable and the explanatory variables under consideration, and this basic assumption can be false if the anthropogenic signals are very strong. For example, it is easy to see that the relationship between load of NH<sub>4</sub>-N and water temperature in the Elbe changed significantly after the reunification of Germany. Second, we noted that the Root Mean PRESS values were often considerably larger than the standard deviation of the residuals, which means that over-fitting can make the time series of normalized values unrealistically smooth. This is not a major problem if the purpose of normalization is to produce point estimates of anthropogenic effects, but it does indicate that caution should be observed when interpreting the results of statistical trend tests applied to normalized data.

#### 6.2 Extracted anthropogenic signals

Data collected at Schnackenburg upstream of Hamburg have long been used to monitor the contribution from the Elbe Basin to the nutrients exported to the North Sea. The present study showed that anthropogenic signals can also be extracted from data collected downstream of Hamburg in the zone where mixing of sea water and freshwater occurs. In particular, we found that most of the interannual variation in nutrient loads measured at Grauerort and Brunsbüttel could be removed by normalizing these loads with respect to water discharge, salinity, and load of SPM. Further downstream, at Cuxhaven, the mechanisms behind the natural fluctuations were apparently too complex to permit efficient removal of such variation.

Closer examination of the data on point emissions and normalized loads of nutrients at different sampling sites confirmed previous reports that the retention (and transformation) of nutrients in the Elbe River has a strong impact on the export of such substances to the North Sea (Behrendt & Opitz, 1999). This is most obvious for the species PO<sub>4</sub>-P and NH<sub>4</sub>-N which are rapidly taken up by algae. However, the retention is also notable for total-N and total-P. In particular, we found that the decrease in loads of nitrogen and phosphorus in the lower reaches of the Elbe was smaller than the decline in point emissions to the river, and the temporal trends recorded upstream of Hamburg were steeper than the trends further downstream. The latter observation implies that the selection of sampling site is a critical factor when the objective is to determine whether the desired reduction has been achieved. Examination of data collected at Schnackenburg indicates that OSPARs goal of a 50% decrease in nutrient loads will be accomplished: the phosphorus goal has already been achieved, and the normalized loads of total nitrogen are approaching a 50% decline. However, the data collected at Grauerort or Brunsbüttel lead to entirely different conclusions, because they show that the phosphorus goal has not been realized, and there are no indications that the nitrogen goal will be achieved within the near future.

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