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Long term change of nutrient concentrations of rivers discharging in European seas

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ABSTRACT

Cases of severe eutrophication are still observed in European surface waters even though tough regulation has been in place since the beginning of the 1990s to control nutrient losses and inputs in the environment. The purpose of this paper is to evaluate the evolution since 1991 of the quality of the water entering European seas in terms of the concentration of major nutrients (nitrogen and phosphorus), and to analyze the effectiveness of implemented national/international measures and EU legislation in reducing water nutrient pollution. Despite the reduction in large portions of the European territory of agricultural nutrient applications and nutrient point source emissions, the impact on water quality is limited. It is shown using two large river basins that this lack of response for nitrogen, and nitrate in particular, between the reduction of the nitrogen surplus and the recovery of water quality is partly explained by the lag time due to transfer of nitrates in the unsaturated and saturated zones and storage in the soils and aquifers. In order to monitor efficiently the impact of policy implementation on water quality, from that of policy implementations to be able to separate the impact of climate variability from that of policy implementation. It is also recommended to investigate and develop harmonized methodologies for estimating the lag time in order to come up with realistic estimates of response time of water bodies due to the implementation of measures.

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1. Introduction

The impact of nutrient excess on surface water quality and on eutrophication in particular has been documented extensively (EEA, 2001). Severe cases of eutrophication have been reported for the Baltic and North Seas and several coastal lagoons along the shore of the Mediterranean Sea (ICES, 2003; EEA, 2001). The spatial extent of eutrophication ranges from local hot spots in enclosed bays to whole seas such as the Baltic (Artioli et al., 2008).

Protecting European waters is high on the agenda of the European Commission because of their ecological and economical importance. Since the beginning of the 1990s, the European Union has been setting various directives to control and reduce nutrient loads into receiving waters: surface water including transitional and coastal waters, and groundwater. The legislation for controlling eutrophication and nutrient loading into receiving waters is embedded in several pieces of legislation, including the Nitrates Directive (EEC, 1991a), the Urban Waste Water Treatment Directive (EEC, 1991b), the Water Framework Directive (EC, 2000), the Common Agricultural Policy, the Marine Strategy Framework Directive (EC, 2008), as well as Marine Conventions covering all European seas.

After about 20 years of implementation of the Nitrates Directive and the Urban Waste Water Treatment Directive, some improvements of the quality of European surface waters are to be expected. Only a few assessments are available at European scale. The European Environment Agency (EEA, 2009) reported that 35% of surface water monitoring stations from the European network EUROWATERNET showed decreasing trends of nitrate concentration for the period 1992-2005, mainly attributed to the implementation of the Nitrates Directive. Based on the comparison of data from the two Nitrates Directive implementation reporting periods of 1996-1999 and 2000-2003, the European Commission (EC, 2007) indicated that 14% of the surface water stations used to monitor the implementation of the Nitrates Directive exhibited increasing concentrations while 55% had decreasing trends. In its latest report, which refers to the period 2004–2007 (EC, 2010), the European Commission indicates that for fresh surface water, 70% of the monitoring stations show stable or decreasing nitrate concentrations as compared to the period 2004-2007 (EU15). The discrepancy between the EEA (2009) and the EC (2007, 2010) assessments is largely explained by the difference in reference periods, monitoring networks, and methodologies used to compute trends. However, independently from trends, it is important to note that both assessments rely on monitoring points spread throughout the river basins, so the results do not provide an integrated response of the river basins to any actions following policy implementation, but are rather indicative of the local effects.

Few studies have tried to perform assessments at large scale of nutrient riverine discharge in receiving coastal waters. Artioli et al. (2008) evaluated the trends of nutrient budgets for European seas, including nutrient riverine discharge, and linked them with the

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implementation of different policies and management plans. They found that no clear trend could be observed throughout Europe. The same policy resulted in different impacts according to the degree of implementation, local conditions, etc., and also depending on the determinand of interest (nitrogen or phosphorus). For instance, they observed that in the Northern Adriatic Sea nitrogen load from anthropogenic sources has been increasing steadily since 1950, while for phosphorus even though the loads have been halved during the 1975–1985 period, the past accumulation in the sediment hinders any improvement in the water column. Similarly, the authors reported that in the Baltic Proper no significant decreasing trend was detected, despite some drastic reduction in the inland nutrient input.

The difficulty in detecting changes of nutrient loads and concentrations entering the seas is exacerbated by the climate variability, which induces changes in water flow and consequently in nutrient loads. This has been observed for the North Sea (Radach and Pätsch, 2007) and in the Danube (van Gils et al., 2005).

Lag time due to transfer of nitrates in the unsaturated and saturated zone, as well as delayed effect due to storage in the soil and aquifers is often cited as the reason for the lack of the response in water quality to management actions. Behrendt et al. (2000) estimated the response time of various German river basins to vary from 10 to 30 years, which is similar to the range of 25 to 50 years given by Osenbrück et al. (2006). In chalk river basins in the UK, Jackson et al. (2008) estimated that after a complete stop of inputs from agriculture a period of 40 years is necessary before a significant impact is seen on surface water and a period of 60 years is required to observe changes in the aquifer. The most striking cases of delayed response are those of the Eastern countries that have seen a decrease by half of the nitrogen surplus, but water quality in streams is yet to respond to these changes as large quantities of nitrogen are stored in the soil system and might be released slowly, due to the mineralization process (Grimvall et al., 2000). This was observed by Stålnacke et al. (2004) in rivers in Estonia, Latvia and Hungary. Similarly, for the Nemunas River in Lithuania, Sileika et al. (2006) reported a continuous strong increase of nitrate concentration in surface water due in part to a large storage and accumulation of soil nitrogen during the Soviet period. Research is still needed to understand and quantify at various temporal and spatial scales the processes linking nutrient pressure from anthropogenic sources and the response of catchments in terms of water quality in the context of policy implementation.

The purpose of this article is to evaluate the long term evolution (since 1991) of water quality entering European seas in terms of major nutrients concentrations (nitrogen and phosphorus), and to analyze the effectiveness of implemented national/international measures and European legislation in reducing water nutrient pollution, in particular, for some of the Member States that had to implement the various EU environmental Directives for the longest time. We start by assessing the changes of nutrient pressure originating from human activities for the period 1990-2005, including waste water treatment discharges and fertilizer applications, and by computing the trends (if any) of water quality entering European seas during the 1991-2005 period. Then we investigate the linkages between changes in nutrient pressure and water quality, considering the implementation of national and European regulations and the bio-physical mechanisms affecting the river basin response. We deepen our analysis providing two case studies with different behaviors: the Elbe and the Loire river basins located in Germany and France, respectively. Finally, we discuss the implications of the findings at the larger scale, providing some recommendations.

2. Material and methods

The present study focuses on the quality of the water entering European seas during the last 20 years, and on the efficiency of environmental regulations to reduce water nutrient pollution. In the study, the quality of fresh water discharging into the sea was considered, as it represents an integrated indication of changes occurring at the river basin scale, even though it may not represent the spatial pattern within the river basin itself. However, this is the only true indicator of the response of a river basin to policy implementation, since the response is not affected by the internal distribution and representativeness of the monitoring stations. The analysis covers the period from 1985 to 2005, to detect possible trends, in particular after 1991 when two major pieces of EU legislation controlling nutrient losses to waters entered into force, i.e. the Nitrates Directive and the Urban Waste Water Treatment Directive. The water quality for the years 1985 to 1990 is included in the analysis to illustrate the range of concentrations prior to the implementation of the Nitrates and the Urban Waste Water Treatment Directives.

2.1. Study area

The study area covers all basins draining into European seas. The extent of the area supports a continental wide assessment while maintaining the focus on EU legislation, which affects directly only EU countries. Therefore the area of interest includes EU27, the Balkan countries, Norway, Turkey, and parts of ex Soviet Union Republics. The area was delimited following the physical borders of the main European river basins draining into the Baltic, North, Atlantic, Mediterranean and Black Seas. The resulting total drainage area covers a surface of 5.9×10^6 km². The river basins draining in European seas were derived from the Catchment Database for Continental Europe (Vogt et al., 2007).

2.2. Estimation of nutrient pressure

The nutrient pressures due to anthropogenic activities considered in the study were the inputs from fertilizers used in agriculture (diffuse sources) and those coming from waste water discharges (point sources). To estimate the pressure originating from agriculture, land use maps for the years 1985, 1990, 1995, 2000 and 2005 were developed and combined with nitrogen and phosphorus fertilizer rate applications (Bouraoui et al., 2009). The inputs were then summarized by river basin.

The land use maps were developed using several global databases and sources of information according to the following scheme: 1) the extent of agricultural area and pasture was taken from the HYDE3 database (Klein Goldewijk and Van Drecht, 2006) as 5 mn grids for the years 1980, 1990, 2000 and 2005; 2) the crop types and shares from 1985 to 2005 were derived from the CAPRI database (Britz, 2004) for EU27 and Norway at regional level, and for the Balkans at country level. For the rest of Europe, the land use information at country level was taken from the FAO (FAO, 2009), and was further spatially distributed in Turkey, using information coming from the global land use database SAGE (Ramankutty et al., 2008); 3) the location of arable land was taken from the 1 km grid Global Land Cover for the year 2000 (GLC2000; Bartholomé and Belward, 2005). The crops were then allocated randomly on the arable land, based on the crop share information, at the regional level when possible (EU27, Norway and Turkey) and at national level otherwise. This resulted in 1 km grid land use maps for the years 1985, 1990, 1995, 2000, and 2005; 4) the fertilizer application rates for EU27, Norway and the Balkan region were obtained from CAPRI (Britz, 2004) and covered the period 1985–2005. They were available as application rate of mineral and manure nitrogen and phosphorus per crop and grassland. For the remaining countries, fertilization rates were taken from the FAO (2002). The fertilization rates were combined with the spatial land use information to derive a total fertilizer application map at 1 km resolution for the years 1985, 1990, 1995, 2000 and 2005.

To estimate nutrient sources coming from waste water discharges, information on population density was combined with national rate of connection to waste water treatment plants and level of nutrient abatement (Bouraoui et al., 2009). The location of urban areas was retrieved from GLC2000 (Bartholomé and Belward, 2005) and was linked to population density (Klein Goldewijk and Van Drecht, 2006) for the years 1985, 1990, 1995 2000, and 2005 to derive a 1 km grid of population count classified either as urban or rural. This information was then combined with human emission factors, national connection level to waste water treatment plants and level of treatment for the respective years available from EUROSTAT, OECD, EEA, and WHO (for more details see Bouraoui et al., 2011). This resulted in point source emissions maps at 1 km resolution for the years 1985, 1990, 1995, 2000 and 2005.

Diffuse and point inputs of nutrients initially available as 1 km grids, were aggregated at the river basin level for the years 1990, 1995, 2000 and 2005. No annual estimation of point and diffuse inputs was performed between each five year time slots as the extent of agricultural area and the population count were only available every five to ten years (Klein Goldewijk and Van Drecht, 2006). The total change in nutrient inputs from 1990 to 2005 (input in 2005–input in 1990) was calculated for each river basin, with a positive change (larger than 10%) indicating an increase of pressure and a negative change (smaller than -10%) a decrease. The sign of the change provides an overall assessment of the situation in 2005 compared to that of 1990, considered as the baseline year.

To dispose of longer time series of agricultural nutrient pressure on a continuous annual basis, national soil surface nutrient balances were calculated. In the case of nitrogen the balance was computed as follows:

$$N_{Bal} = N_{Fert} + N_{Man} + N_{Dep} + N_{Fix_{symb}} + N_{Fix_{asymb}} - N_{Upt}$$
(1)

where N_{Bal} is the nitrogen balance (also called surplus), N_{Fert} is the mineral nitrogen fertilizer application, N_{Man} is the manure application, N_{Dep} is the nitrogen deposition, N_{Fixsymb} is the symbiotic nitrogen fixation (occurring in plants), $N_{\ensuremath{\text{Fixasymb}}}$ is the asymbiotic nitrogen fixation (occurring in soils), and N_{Upt} is the nitrogen uptake and all units are in kg N/ha. For phosphorus the balance was computed using a similar equation, however no phosphorus deposition was considered. The balance was computed annually from 1965 to 2005 at country level. The total nitrogen and phosphorus fertilizer consumptions were retrieved from FAOSTAT (FAO, 2009) and used unmodified. The head stocks of ducks, chicken, buffaloes, cattle, pigs, geese, and horses were also retrieved from the FAOSTAT and were then converted in manure production using excretion coefficients (Bouraoui et al., 2009). The wet and dry nitrogen atmospheric deposition data were obtained from the Cooperative Programme for the Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP, 2001).

2.3. Water quality data and trend analysis

To perform the trend analysis on water quality parameters, data on nutrient (nitrogen and phosphorus) concentrations at the basins outlets were retrieved from OECD (2008). These data were chosen for the study as they are officially reported by member countries to OECD, they have a wide spatial extent, and they cover the period of interest (1991–2005). Furthermore, their extent is long enough to be able to separate short term fluctuation mostly controlled by climate variability from long term trend due to a change in the system's behavior (for example Howden et al. (2011) reports a minimum period length of twelve years in the UK). From the original 77 stations available from OECD (2008), only the ones corresponding to a river basin outlet to European seas were kept, resulting in 39 stations with annual measurements of nitrate (NO_3) , ammonium (NH₄), and total phosphorus (TotP) concentrations. To extend the spatial coverage of the analysis, annual data for the Baltic and the Danube were retrieved from other sources. For the Baltic area, we used data of dissolved inorganic nitrogen (DIN) and TotP concentrations available from HELCOM (2009) and described in Mörth et al. (2007), resulting in an additional 51 stations. Flow data were collected according to the guidelines of the World Meteorological Organization and water quality measurements were collected at least on a monthly basis and are representative of the flow (HELCOM, 2004). Data analysis for nitrogen and phosphorus was performed using a similar method for most of the contracting parties (HELCOM, 2004). The data consisted in monthly water quality load and water discharge that were combined to obtain an average annual flow weighted concentration of DIN and TotP. Similar monthly data were obtained for the Danube (ICPDR, 2009) to compute average annual flow weighted concentrations of DIN and TotP. In total, within the target period 1991–2005, 90 stations were available for DIN and TotP (HELCOM + ICDPR + OECD stations), and 39 stations for NO3 and NH4 (OECD stations).

The trend analysis was performed using the non-parametric test of Mann-Kendall (Hirsh et al., 1991). This test does not make any assumption regarding the data distribution and deals with incomplete, seasonal data with serial dependence, and any type of trend (linear and non-linear). The first step of the test is to determine the sign of the n(n-1)/2 differences between the pairs $(x_j;x_k)$ with j < k and to compute the Mann-Kendall S with the following convention:

$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^{n} s(x_j - x_k) \text{ where } \begin{cases} s(x_j - x_k) = 1 & \text{ if } x_j - x_k > 0; \\ s(x_j - x_k) = 0 & \text{ if } x_j - x_k = 0; \\ s(x_j - x_k) = -1 & \text{ if } x_j - x_k < 0. \end{cases}$$
(2)

where $x_1,...,x_n$ are the water quality variables ordered in a chronological way, and n is the number of points (years) to be analyzed. The probabilities for the Mann-Kendall non parametric test for trend for n less than 40 are given in Hollander and Wolfe (1973). For large data sets (n <= 40), the Z test statistics is then computed. Additional details about the test can be found in Gilbert (1987). Trends were tested at a 90% level of significance.

2.4. Analysis of the Elbe and Loire river basins

To better understand the processes controlling the catchment response to variations of nutrient pressures, and especially to improve the understanding of the physical mechanisms underpinning the river basins response to input changes, two cases with different behaviors, including the Elbe and Loire river basins located in Germany and France, respectively, were analyzed. The Elbe extends over an area of 148×10^3 km², mainly in Germany. It receives contributions also from Czech Republic, Poland and Austria. The Elbe has a population of about $25\!\times\!10^6$ inhabitants and around 50% of the basin is dedicated to agriculture. The Loire covers an area of 118×10^3 km². Around 65% of the area is occupied by arable land and the human population is around 12×10^6 inhabitants. The analysis focused exclusively on nitrates, which are a good indicator of agricultural pressures as they easily percolate to aquifers or reach surface water via drains or surface runoff. The data for the Elbe was retrieved from Radach and Pätsch (2007) and consisted of daily nitrate fluxes and water flow for the period 1977-2006. For the Loire, water quality data was retrieved from the Loire-Bretagne Water Agency and consisted in monthly measurements of nitrate concentration, while daily water flow was obtained from the French Ministry of Ecology and Sustainable Development. Both data sets covered the period 1977-2005.

The nitrate concentration of the two components of river discharge, baseflow and surface runoff, was estimated and cross-correlated to the long term national nitrogen balance. Baseflow is the portion of the water in the stream that is not coming from direct surface runoff and is usually largely sustained by groundwater discharge in the stream. To separate the total flow of the Elbe and Loire river basins into surface runoff and baseflow, we used the recursive digital filter developed by Arnold and Allen (1999). The software LOADEST (Runkel et al., 2004) was used to compute daily nitrate loads and concentrations in surface runoff and baseflow. In LOADEST, the concentrations are derived from a sitespecific regression based on the relationship between concentration and river discharge:

$$Ln(Nl) = a0 + a1 lnQ + a2 lnQ^{2} + a3 sin(2 \pi dt)$$
(3)
+ a4 cos(2 \pi dt) + a5 dt + a6 dt2

where NI is the nitrate load in kg N/day, lnQ is the centered natural logarithm of the flow (m^3/s), dt is the centered decimal time, and a0, a1, a2, a3, a4, a5 and a6 are site specific regression coefficients.

The procedure described by Schilling and Zhang (2004) was used to calculate the nitrate concentration (and load) in the baseflow and surface runoff. For the Loire river basin LOADEST was used to extrapolate the monthly nitrate concentration measurements to daily concentrations based on the daily flow measurement according Eq. (3). When the baseflow represented at least 90% of the total flow, it was assumed that the nitrate concentration on that specific day was equal to that of the baseflow. LOADEST was then run again to estimate the baseflow nitrate concentration (and load) using the baseflow value (Q) as explanatory variable in Eq. (3). For the Elbe, daily loads were already available, so in this case LOADEST was used only once to estimate the nitrate concentration (and load) in the baseflow as described in the case of the Loire.

The nitrate concentration was then cross-correlated to the national nitrogen surplus in France and Germany calculated according to Eq. (1). The cross correlation was used to detect how long it would take to see an impact of a change in nitrogen surplus on the baseflow nitrate concentration. The national surplus was used as it is the only available continuous indicator of the change of agricultural pressure on an annual basis (the pressures at the river basin level were available by time segments of five years and could not be used for a cross correlation analysis). Cross-correlations were tested at a 90% level of significance and were computed with the software SPSS (2003).

3. Results and discussion

3.1. Changes in nutrient pressure

The change in nutrient pressure for the whole Europe was analyzed, with a particular focus on EU15 (see list of countries in Table 1) whose countries have been implementing the Nitrates Directive and the Urban Waste Water Treatment Directive for the longest time. The nitrogen balance for the time period 1960–2005 for all the 39 countries with river basins draining in European seas is shown in Fig. 1. In 2005, the highest

Table 1

Change of anthropogenic nitrogen pressure for EU15 between 1990 and 2005.

	Point source nitrogen 1000 tons		Manure nitrogen 1000 tons		Mineral nitrogen 1000 tons	
	1990	2005	1990	2005	1990	2005
Finland	4	4	56	55	199	175
Sweden	10	10	142	161	215	202
UK	184	208	1180	1044	1541	1113
Denmark	17	8	265	352	382	229
Ireland	12	15	466	426	390	349
The Netherlands	55	23	455	477	392	284
Luxembourg	1	2	10	9	13	9
Belgium	9	32	224	251	160	150
Germany	191	114	1359	1375	1820	1786
Austria	21	10	147	174	137	116
France	165	131	1267	1452	2495	2283
Italy	230	218	642	769	752	753
Greece	24	28	268	219	592	268
Portugal	27	26	141	146	144	88
Spain	100	222	755	1211	937	1192
Total	1047	1051	7378	8121	10169	8997

nutrient surpluses are found in The Netherlands and Belgium. Most of the countries in Western Europe show a decrease in the nitrogen surplus after the increase registered during the 1960s and the 1970s. Among EU15 countries, only Spain has seen a constant increase of the nitrogen surplus. A large part of the nitrogen surplus reduction in Western Europe is due to the combination of several factors including 1) the reform to limit milk production and control production surplus in the 1980s in order to accommodate the new Members joining the EU, 2) the introduction of set aside, 3) the increase use of fixing crop, 4) the rise of the price of fertilizers, and 5) the increase of nitrogen use efficiency due to better management practices and better selection of crop variety used (Eickhout et al., 2006). In Eastern European countries formerly part of the ex Soviet Republics, after a sharp drop in 1990 nitrogen surpluses are showing an upward trend indicating a pick up in the agricultural activities.

The increase of nitrogen use efficiency of crops is illustrated by the French case (Fig. 1). Even though the surplus has been declining since the late 1980s, the amount of applied nitrogen has decreased only starting from 2000, without any decline in the production, indicating a more efficient nitrogen crop uptake. It is interesting to note that for several countries (Denmark, The Netherlands, UK, and Germany) the drop in the nitrogen surplus has started before 1991, date of adoption of the Nitrates Directive. This indicates that national regulation aiming at controlling nitrogen losses combined with better agricultural practices improving crop uptake were in place prior to the Nitrates Directive. For instance in Denmark the control of nitrogen surplus started in the mid 1980s, simultaneously with the implementation of the 'Action Plan I on the Aquatic Environment' mandated by the Danish government to reduce nitrogen leaching from agriculture (Ambus et al., 2001).

Fig. 2 displays the national phosphorus balances for the time period 1960-2005 for the 39 countries of the study area. In many cases, the decrease in the phosphorus surplus started much earlier than that of nitrogen and was not based on environmental regulation (for Denmark see Kyllingsbæk and Hansen, 2007) but rather related to the enrichment of phosphorus in agricultural soils. In fact, this decrease occurred because phosphorus was no longer a limiting factor to optimum crop yield, while the same condition was reached later for nitrogen. For some countries, like The Netherlands, the drop in phosphorus surplus started at a later stage than in Germany. Indeed, in Germany there was a contemporary decrease of the application of mineral phosphorus and the production of manure phosphorus, while in the Netherlands the production of organic phosphorus through animal waste continued to increase until around 1995, despite a huge continuous decline of the amount of phosphorus applied as mineral fertilizer since the mid 1960s. It is important to stress that the reduction of phosphorus surplus is not due to the implementation of environmental regulations as in Europe only a limited number of countries have legislation limiting directly the amount of applied phosphorus, including The Netherlands, Ireland, Norway and Sweden (for more details see De Clercq et al., 2001).

The change in fertilizer application per river basin in 2005 with respect to the baseline condition of 1990 is shown in Figs. 3 and 4 for nitrogen and phosphorus, respectively. Spain exhibits an increase of nitrogen and phosphorus application throughout the country. The larger river basins in Western Europe that show an increase between 1990 and 2005 of nitrogen application include the Loire and the Rhone in France, the Po in Italy, and the Weser in Germany. Concerning phosphorus, only Spain exhibits an increase of application throughout its territory. For the rest of Europe, the large majority of the river basins have seen a decrease of phosphorus application.

A similar analysis was conducted for the change in point source inputs of nitrogen and phosphorus per river basin between 1990 and 2005 (Fig. 5). Point source emissions of nitrogen and phosphorus have decreased in large portion of Europe, except in England and Spain. For England this estimated increase is partially due to the lack of information available about the change in time of the level of connection of people to waste water treatment plant and the level of treatment, so a unique



Fig. 1. National nitrogen balances (kg N/ha UAA) for the period 1960–2005. The vertical reference line indicates the year of implementation of the Nitrates Directive.



Fig. 2. National phosphorus balances (kg P/ha UAA) for the period 1960–2005. The vertical reference line indicates the year of implementation of the Nitrates Directive.



Fig. 3. Change of nitrogen fertilizer application between 1990 and 2005 at river basin level (input in 2005-input in 1990).

value was used throughout the study period, and the increase of point source emission reflects here the increase of population.

Globally, for EU15, the nitrogen surplus has decreased by about 32%, from 9.6×10^6 tons of nitrogen to 6.6×10^6 tons of nitrogen between 1990 and 2005. The amount of applied fertilizer (mineral, organic, and crop fixation) has decreased by 13%, so large part of the decrease of the nitrogen surplus is explained by increased nitrogen use efficiency. During the same time period the point source input of nitrogen for EU15 has remained stable around 1.0×10^6 tons of nitrogen however with large differences from one country to the other. For instance Germany has seen a decrease of the amount of nitrogen from point sources by 40%, the Netherlands by 60%, while Spain has seen an increase by 100% of nitrogen emissions from point sources resulting from an increase of the connection of the population to water treatment plants (the connection rate increased from around 42% in 1990 to 100% in 2005). It is important to note that in 2005, the nitrogen surplus for EU15 is about six times larger than point source emissions of nitrogen. A summary for the changes in nitrogen anthropogenic pressure for EU15 is given in Table 1.

To analyze if these decreases in nutrient inputs observed in many countries had any effect on water quality at the river basin outlets, a trend analysis was performed for various determinands, including, nitrate, ammonium, dissolved inorganic N, and total phosphorus.

3.2. Trends analysis of water quality

When possible, data from the OECD was cross checked in terms of relative evolution to other data sources described in Bouraoui et al. (2009). This cross comparison was made for the Po River and all French and German rivers, and no discrepancy was found in terms of trend.

The long term nitrate concentrations for the OECD monitoring stations located at the basins outlets are shown in Fig. 6 and the results of the trend analysis using the Mann-Kendall test are displayed in Fig. 7. About 30% of the monitoring stations show a decreasing trend and 10% an increasing trend. Most of the major river basins draining in the North Sea exhibit a decreasing nitrate concentration trend, more marked in Germany, in particular for the Elbe, Rhine, and Weser (Fig. 6). A similar rapid decrease is observed for the Odense catchment. In the Mersey river basin, which discharges in one of the most heavily polluted estuaries in Europe (Burton, 2003) the nitrate concentration has jumped from 3 mg N/L in 1991 to around 6 mg N/L in 2004, due to the contribution of both agriculture and point sources (Rothwell et al., 2010). This dependence of the nitrate concentration on the contribution of both point and diffuse sources is found in other river basins and was also confirmed by a correlation analysis (Table 2). A significant correlation is found between nitrate concentration and the pressure from point sources (expressed as total nitrogen emission per unit area) and from diffuse sources (expressed as total nitrogen fertilizer applied per unit area). Even though some basins exhibit a decreasing nitrate trend since 1991, the concentration at the outlet remains high, such as the case of the Thames, where nitrate concentration in 2003 was above 7 mg N/L. The results for the UK rivers (Thames and Severn) are consistent with those found by Burt et al. (2011) who report that the nitrate concentrations in many rivers have leveled off. However the concentrations in the UK rivers are the highest among the OECD



Fig. 4. Change of phosphorus fertilizer application between 1990 and 2005 at river basin level (input in 2005-input in 1990).

stations. One can note the hectic behavior of the nitrate concentration in the Guadalquivir. It must be noted that after 1990 there was a change of the monitoring station. In fact the original station was replaced by a station closer to the river mouth away from the influence of the town of Seville. However, it seems that the nitrate concentration might be under a tidal influence.

When computing the trend of nitrate concentration as the sign of the difference between the two consecutive reporting periods 1996–1999 and 2000–2003, according to the approach used in the evaluation of implementation of the Nitrates Directive (see EC, 2007), it results that 67% of the monitoring stations have a decreasing trend (55% reported by EC, 2007), which is much higher than the value obtained by the nonparametric trend analysis (30%). For this reason it is suggested that assessing the changes in water quality by comparing two different periods could not provide a realistic indication of the true trend of concentration. In fact, such an approach gives only a snapshot of difference of concentrations between two time periods, and could be strongly biased by climatic variability. We found a significant (0.05 level) correlation between the annual nitrate concentration and annual precipitation for 10 stations (out of 39), 8 of which exhibit positive correlation. This clearly indicates that for those stations nitrate concentration will tend to be lower during dryer years and higher during wet years.

As for nitrate, trend analysis of ammonium concentration was performed for the 39 monitoring stations available. The time series of ammonium concentrations and corresponding trend analysis are shown in Figs. 8 and 9, respectively. About 8% (3 stations) of the stations have an increasing trend and 36% a decreasing trend. The increasing trends for ammonium are limited to very small basins located in Greece, Italy and Sweden. Larger areas are affected by a decrease in ammonium concentration than by a decrease of nitrate concentration. Like for nitrate, the German catchments exhibit a very marked decreasing ammonium trend, illustrating the improvement of waste water treatment that has been taking place in Germany for the past twenty years. The Odense catchment is also exhibiting a continuous decrease of ammonium concentration since 1991, but also prior to that date. The Mersey catchment is characterized by a decreasing ammonium trend, illustrating the effort to control and remove point pollution of industrial and urban effluent discharging in the river (Burton, 2003). We predict a decreasing trend of ammonium concentration in the Seine (France), highlighting the continuous efforts to control waste water effluents, in particular those from the city of Paris (Billen et al., 2001). Similarly the decreasing trend of ammonium predicted in the Ebro highlights the efforts that started in the late 1980s and early 1990s to reduce the pollution load coming from waste water treatment plants in Spain, in particular in the Mediterranean region near the tourist areas (Kemp, 2001). The dependence of ammonium concentration on the pressure from point sources is confirmed by the correlation analysis (Table 2). Indeed ammonium concentration is strongly linked to emission from point sources and no correlation was found with diffuse sources. For all basins, ammonium concentrations are much lower than those of nitrates







Fig. 6. Time series of the nitrate concentration for the OECD stations. The vertical reference line indicates the year of implementation of the Nitrates Directive.



Fig. 7. Trend analysis of the nitrate concentration for the 39 river basins analyzed for the period 1991–2004.

The trend analysis of the dissolved inorganic nitrogen concentrations for all available stations is shown in Fig. 10. We estimated that 12% of the stations have an increasing trend while 23% a decreasing trend. Performing the same analysis using only the OECD stations (39) resulted in 13% of the stations having an increasing trend and 39% a decreasing trend. The large difference highlights how important is the selection of the monitoring network. Indeed the majority of the stations of the Baltic region have rather low DIN concentrations and exhibit no trend. It is interesting to note the increasing trends of DIN in certain parts of Turkey, Greece and Spain which can be explained by the intensification of agriculture, and increased pressure from point sources (Figs. 3 and 5). Indeed the correlation analysis shows an equally strong relationship between DIN concentration and point emissions and diffuses sources (Table 2). We can note also an increase of DIN in four Finnish catchments all draining in the Bothnian Bay. This increase, mostly controlled by the trend of nitrate, was also reported by Räike et al. (2003) and is explained by the intensive agriculture and also the

Table 2

Pearson correlation coefficients between nitrate, ammonium, DIN, total P concentrations and point and diffuse sources (N or P depending and the water quality parameter considered) for year 2005. Both point and diffuse sources are normalized by the area of the river basins.

	TOT P	DIN	NO3	NH4
Diffuse sources	0.33 ^{**}	0.69^{**} 0.69^{**}	0.63 ^{**}	0.05
Point sources	0.50 ^{**}		0.76 ^{**}	0.77 ^{**}

** Correlation is significant at the 0.01 level.

presence of peatland. It is important to stress that ammonium concentrations have dropped drastically in most Finnish rivers (Räike et al., 2003), so the predicted increasing trend of DIN is controlled mostly by the increase of nitrate concentrations. A similar case is also observed for the Loire (France), the Duero (Spain and Portugal), where the increasing trend of DIN is controlled by the increasing trend of nitrate, largely due to the intensification of agriculture (Fig. 3).

The three major catchments draining in the Mediterranean exhibit no trend (Po) or even a decreasing trend (Ebro and Rhone) for DIN. The Ebro decreasing trend is mostly controlled by the decrease of ammonium, also reported by Ibáñez et al. (2008) while for the Rhone there is a decreasing trend for both nitrate and ammonium reported also by Olivier et al. (2009). For the rivers discharging in the Baltic, DIN concentrations are in most cases decreasing or not exhibiting any trend with the exception of four Finnish catchments located in an intensive agricultural area. Most of the rivers draining in the North Sea exhibit a decreasing trend of DIN.

The time series of total phosphorus concentration are shown in Fig. 11. The results for trend analysis for total phosphorus are shown in Fig. 12. It has been estimated that 8% of the stations have an increasing trend and 38% a decreasing trend (3% and 32% when using only the OECD stations). Total phosphorus concentration exhibits a strong significant correlation with point source emission. Total phosphorus also has a significant positive correlation with diffuse sources, but weaker than that for point sources. Most of the Baltic stations are characterized by a decrease of total phosphorus concentrations, highlighting the improvement of phosphorus removal in waste water treatment plants. Similarly,



Fig. 8. Time series of the ammonium concentration for the OECD stations. The vertical reference line indicates the year of implementation of the Nitrates Directive.



Fig. 9. Trend analysis of the ammonium concentration for the 39 river basins analyzed for the period 1991–2004.

most of the German river basins, except the Rhine, exhibit a decreasing trend. Fig. 11 shows that there was a sharp decrease in the total phosphorus concentration in the Rhine river basins before 1991, remaining stable afterwards. For the other river basins the decrease of total phosphorus takes place over a longer time period showing the impact of the regional difference in efficiency in phosphorus removal and waste water treatment plants implementation. For the Western Dvina (Latvia) there is an increasing trend of total phosphorus mostly controlled by the increasing incoming phosphorus from urban and industries from Russia whose headwaters control the chemistry of the river (Henn et al., 2009). However most of the catchments exhibit a decreasing trend or no trend highlighting the development in time of higher phosphorus removal in waste water and also the positive impacts of the ban in several countries of phosphates and other phosphorus compounds in household laundry detergents.

3.3. The cases of Elbe and Loire

Our results show than even though a large portion of the European territory has seen for various reasons significant decreases of the amount nitrogen and phosphorus entering directly or indirectly surface waters, there are still many catchments where the response to these changes is not evident. Furthermore, there are also still areas in Europe, including countries of EU15, where nitrate concentrations in surface waters are increasing despite stringent regulation controlling nitrate losses. To better understand the phenomenon controlling the response of catchments to large changes in nutrient pressures, we analyzed two case studies: the Elbe where there is a decreasing trend of nitrate and a decrease of nitrogen surplus (Fig. 1); and the Loire where there is an increase of nitrate concentration and a decrease of nitrogen surplus (Fig. 1).

The application of the baseflow separation for the Elbe river basin is shown in Fig. 13. In 2005, baseflow contributes 75% of the total runoff and 63% of the total load of nitrate. In the Elbe there seems to be a dilution effect from the groundwater. Indeed when calculating the concentration of the various components of the runoff (Fig. 13), it is clear that the nitrate concentration in surface runoff is higher than in the baseflow (groundwater). It is also interesting to note the decrease in the baseflow nitrate concentration starting from the late 1980s. A cross correlation was performed between the baseflow concentration and the national German nitrogen surplus, and the highest positive significant correlation (R=0.54) indicates a lag time of 8 years between the nitrogen surplus and the concentration in the baseflow. This means that in the current situation 8 years would be necessary before the variation in nitrogen surplus affects the nitrogen concentration in the baseflow and the shallow aquifer in general. No significant correlation was found between the nitrogen surplus and the nitrate concentration in surface water.

Concerning the Loire, in 2005, the baseflow contributes 75% of the total flow and 83% of the total nitrogen load (Fig. 14). Unlike the Elbe, in the Loire there is enrichment of nitrate in surface water, due to the groundwater contribution. Indeed, an analysis of the concentration of the various components of runoff shows a higher concentration in baseflow than in surface runoff (4.7 and 3.4 mg N/L, respectively).



Fig. 10. Trend analysis of the dissolved inorganic nitrogen concentration for the 90 river basins analyzed for the period 1991–2004.

Starting in 1990, there is an increase on the nitrate concentration in groundwater from 2 mg N/L to 4.7 mg N/L in 2005, while in the same time there seems to be less variability in the nitrate concentration in surface runoff. The results of the cross correlation between the French national nitrogen surplus and the baseflow concentration reveal a maximum significant correlation coefficient of 0.46 with a delay of 14 years. This indicates that the increase of the nitrate concentration in the baseflow is affected by nitrogen surpluses that have occurred 14 years ago. The pattern of the nitrate concentration in the baseflow is characteristic of breakthrough curve that have been documented in literature (Howden and Burt, 2008). Considering that in the Loire nitrogen application have increased until at least 2005, nitrate concentration in groundwater is likely to increase in the decades to come.

The two cases of Elbe and Loire highlight the importance to consider the impact of lag time when analyzing water quality time series in order to understand what type of measures are the most effective in controlling nitrate losses and the lag before the effects of these measures can be detected.

Concerning the Elbe, the decrease in the nitrate concentration (total and baseflow) started before the implementation of the Nitrates and Urban Waste Water Directives. The analysis also indicated that any change in nitrogen surplus would have taken about 8 years before any effects were seen. So the improvement of the water quality is in part due to the efforts that were taken before the implementation of these two directives. The implementation of the Directives then reinforced or maintained the decreasing trend. Similar evidence was found in the Odense river basin in Denmark, where the decrease of nitrate concentration started in the late 1980s as a result of a stricter environmental policy put in place before the Nitrates and Urban Waste Water Directives (Ambus et al., 2001). Differently, in the Loire there is an increase of the nitrogen application, which combined with the past overfertilization is leading to a continuous degradation of the ground water quality. This phenomenon is expected to continue for at least 14 more years due to the inertia of the system to respond to changes.

It is clear that the countries where the implementation of the Nitrates Directive and Urban Waste Water Directive is lagging behind are unlikely to see immediate (any) improvement in the water quality. It is also possible due to the delay effects that were just highlighted, that some peaks of nitrates will occur in the future due to past management strategies. So it is impossible without a detailed analysis to conclude that a no trend of nitrate concentration in a specific monitoring stations is a good sign of water quality (quality improvement) as delays up to 40 years between fertilizer application and nitrate concentrations have also been documented (Jackson et al., 2008).

3.4. Impacts of EU regulations

Two major European Environmental Directives affecting nutrient quantity in water bodies have been in place for about 20 years. A clear impact on the nutrient pressure side has been noted for several countries concerning both the input of point sources and fertilizer application.



Fig. 11. Time series of the total phosphorus concentration for the OECD stations. The vertical reference line indicates the year of implementation of the Nitrates Directive.



Fig. 12. Trend analysis of the total phosphorus concentration for the 90 river basins analyzed for the period 1991–2004.

However full implementation of the Nitrates and the Urban Waste Water Treatment Directives has not been achieved and infringement cases of the Nitrates Directive are reported for several countries of EU15 including France, Luxembourg and Spain (EC, 2010). Concerning the Urban Waste Water Treatment Directive, the European Commission states that 'secondary treatment needs to be improved in some EU15 Member States', and that 'compliance rates for more stringent treatment are very low in some EU15 countries and, overall greater efforts in implementation are needed' (EC, 2009). Where environmental legislation was already in place before 1991, the Directives have helped maintaining and improving the momentum of decrease of nutrient input. In other cases, the Directives, when implemented properly, have limited the amount of nutrients entering water bodies. However some countries are lagging behind in implementing the legislation and the improvement in water quality is yet to be seen. The positive effects of legislation and measures on water quality are also hampered by the delayed response of the environment to external changes. It was shown that 8 years and 14 years are the time expected for the Elbe and Loire



Fig. 13. Baseflow and load separation (left panel) and nitrate concentration in the various components of the runoff (right panel) for the Elbe river basin.



Fig. 14. Baseflow and load separation (left panel) and nitrate concentration in the various components of the runoff (for the Loire river basin right panel).

rivers to react to changes in nutrient application, respectively. Longer response times have been reported for other German and English river basins (Behrendt et al., 2000; Jackson et al., 2008).

An additional difficulty in evaluating the effectiveness of legislation and/or measures implemented is the choice of the right metrics to evaluate response to changes, both in terms of indicators, and spatial and temporal scales. Indeed, choosing the national nitrogen surplus for instance as an indicator, one can conclude that many countries are having the good strategy in controlling nutrient losses. However this indicator hides the large spatial variability which is observed within some countries. For instance France is exhibiting a steady decrease of the nitrogen surplus; however it has been shown that the Loire, the largest French river basin has an increasing nitrate concentration at the outlet.

In addition, this study showed that characterizing a improvement (or degradation) of water quality as the difference in concentration between two time periods can lead to a different outcome to that obtained computing the trend using long term time series, since nitrate concentrations are in many cases climate dependent. Indeed, trends should be evaluated over the long term in order to separate out the impacts of climate. So the reporting mechanism from Member States to the Commission should focus on getting long term water quality concentration on a permanent network of monitoring stations located as close as possible to the river basins outlets. Burt et al. (2011) also advocated maintaining long term permanent monitoring systems in order to evaluate the impacts of policy and management options. Permanent monitoring stations are also required within the river basins to evaluate the local implementation of measures, but the true integrated response of the river basin indicating if further efforts are needed to reduce nutrient concentration can only be measured at the outlet. Efforts should also focus on harmonized approaches to characterize the hydrogeological response of river basins and in particular the lag time, in order to quantify the response of aquifers to changes in legislation or implementation of measures, and avoid erroneous conclusions. A simple method was illustrated here on how to calculate the lag time without having to undergo lengthy hydrogeological surveys and complex modeling. Such an approach could be implemented at large scales.

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Konklusion på Europakommissionens rapport vedrørende effekten af de politiske tiltag man i EU siden 1991 har foretaget for at forbedre kvaliteten af vandmiljøet ved at begrænse udledning af kvælstof fra landbrug og byer.

Faycal Bouraoui og Bruna Grizzetti har i rapporten "Long term change of nutrient concentrations of rivers discharging in European seas" (Ændringer over længere tid i koncentrationerne af næringsstoffer i floder med udløb i europæiske have) undersøgt, hvad resultatet har været af de tiltag, som en række EU-lande har foretaget for at reducere udledningen af fosfor og kvælstof i det europæiske vandmiljø.

Rapporten er bestilt af Joint Research Centre of the European Commission, og den er godkendt af EU Kommissionen den 9. august 2011.

Ved at sammenligne indholdet af fosfor og kvælstof i overfladevandet i en række europæiske vandløb over en 20 års periode (1985 – 2005), heriblandt tre danske, nemlig; Gudenåen, Skjern Å og Odense Å har forskerne konkluderet, at selv om man reducerer forbruget af gødning i landbruget i stor skala, samtidig med at man har fjernet en stor del af byernes punktforurening med næringsstoffer, så har en mindsket kvælstofudledning en meget begrænset effekt på vandkvaliteten af overfladevandet i de indre europæiske, og danske, farvande.

I et nærstudie af Loire-floden fandt man, at på trods af et fald i udledningen af kvælstof over en lang årrække, så steg kvælstofmængden i floden, og i Elben fandt man, at der var et fald i mængden af kvælstof i floden samtidig med et fald i udledningen af kvælstof. Rapporten konkluderer, at en begrænset kvælstofudledning ikke har en signifikant effekt på kvælstofmængden ved flodudmundingerne. Det konkluderes, at målingerne af kvælstofs effekt på vandmiljøet skal foretages over en meget lang periode, da effekten af en meget varm sommer er langt større end effekten af at begrænse kvælstofudledningen. Oplever man derimod en meget våd sommer, så vil udvaskningen fra jorden være større end normalt, og man vil altså opleve en stigning i kvælstofmængden i vandmiljøet – også selvom der er blevet udledt mindre kvælstof fra landbruget og byerne.

Med EU-Kommissionens rapport i hånden må man konkludere, at de bestræbelser Danmark siden 1987 har gjort sig for at begrænse udledningen af kvælstof, har haft en meget begrænset effekt på kvaliteten af vandmiljøet i Danmark.

Af Jakob Tilma, Bæredygtigt Landbrug