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A model reconstruction of riverine nutrient fluxes and eutrophication in the Belgian Coastal Zone since 1984

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ABSTRACT

The OSPAR convention signed in 1992 by 15 European states including Belgium and France pledged to reduce the nutrient (nitrogen N and phosphorus P) loads from land-based sources to the Channel and the North Sea to half of what they were in 1985. In this paper, we use a river basin–coastal sea chain model to describe the evolution of nutrient loads to the Belgian Costal Zone originating from the Seine, Somme and Scheldt watersheds from 1984 to 2007 in order to assess the N and P reduction with respect to the OSPAR goals and the resulting effect on coastal eutrophication, especially *Phaeocystis* blooms. Since the early 1990s, most nutrient reduction actions have been devoted to domestic and industrial wastewater treatment, resulting in a sharp P decrease between 1984 and 2007: from 260 to 90 kgP km^{-2} for the Seine River and from 215 to 110 kgP km^{-2} for the Scheldt River. In spite of improved N treatment of wastewater, there is no clear decrease of N loads, which mostly originate from leaching intensively cultivated arable lands. N fluxes at the outlet of the Seine and Scheldt rivers were, respectively, 1990 and 2210 kgN km^{-2} in 1984 and 1830 and 1390 kgN km^{-2} in 2007. However, this relatively low decrease appears to be more influenced by hydrological conditions than by better efficiency of N use in agriculture. We conclude from this analysis that the OSPAR objectives for P have been achieved, whereas for N radical changes in agricultural practices are still required. The P reduction achieved allows, for the period of concern, a 50% decrease of *Phaeocystis* colony blooms in the Belgian Coastal Zone, both in magnitude and duration. However, the simulated decrease, of maximum abundance, i.e., from $60 \cdot 10^6$ in 1984 to $30 \cdot 10^6$ cells l^{-1} in 2007, is still insufficient when compared to the ecological-quality indicator of $4 \cdot 10^6$ cells l^{-1}. A further decrease of nutrients is still necessary to decrease undesirable blooms more satisfactorily.

1. Introduction

Coastal eutrophication, following that of freshwater lake systems (Vollenweider, 1969), has been one of the most challenging environmental issues since the 1980s and remains so at the beginning of the 21st century. The imbalance in river inputs of nitrogen (N) and phosphorus (P), with respect to those of silica (Si), reflecting human activities within watersheds (Billen et al., 2007a; Galloway et al., 2008; Lassaletta et al., 2009), is the source of non siliceous algal blooms in rivers and coastal waters (Billen et al., 2007b; Howarth and Marino, 2006; Howarth et al., 2011; Officer and Ryther, 1980; Seitzinger et al., 2010). N originates to a large extent from diffuse agricultural sources resulting from an excessive organic or mineral fertilisation of arable land (annual or permanent crops), which has increased in the last 40 years (De Vries et al., 2011). On the other hand, household and industrial wastewater releases still account for the major P sources despite the progress made in water purification and the growing importance of diffuse sources from agricultural soil (Némery and Garnier, 2007).

However, many coastal zones of developed and emerging countries are experiencing severe eutrophication (Diaz and Rosenberg, 2008; Howarth, 2008). Most damaged zones are at the outlet of watersheds with intensive agriculture (Domingues et al., 2011), high population density and inefficient urban wastewater treatment. Eutrophication can lead to hypoxia and even to anoxia as in the Gulf of Mexico (Turner et al., 2005), the Black Sea (Ludwig et al., 2010), China's coastlines (Zhu et al., 2011) and the Baltic Sea (Conley et al., 2011).

In the European Union, although the problem has been pointed out since the late 1960s, at the national scale, citizens, managers and politicians only began to worry about the damage in the middle of the 1970s for stagnant systems and the 1980s for running waters and coastal zones (Ferreira et al., 2011). In 1991, the European Parliament adopted the "Nitrates Directive" (91/676/CEE, 1991a) targeting N pollution

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caused by agricultural activities. Each member state had to identify "vulnerable areas", i.e., where nitrate (NO_3^-) concentration in water was above 50 mgN–NO_3^- l^{-1} or where undesirable eutrophication may occur. Within these areas, "good agricultural practices" must be applied in order to respect the balance between crop nutrient requirements and fertiliser applications (Ruiz-Ramos et al., 2011). Recently, the European Commission referred France to the Court of Justice of the European Union for an excessively relaxed application of these measures. In 1992, the OSPAR convention was signed by bordering NE Atlantic States, who agreed to reduce nutrient export by 50% in 2010 compared to 1985, the year taken as a reference (Foden et al., 2010). In 2000, the "Water Framework Directive" (WFD,; 2000/60/CE, 2000) was adopted by European legislators (Borja et al., 2010). Each member state had to define "hydrological districts" within its territory, in which measures should be taken in order to reach the "Good Ecological Status" (Ferreira et al., 2011) and the "Good Chemical Status" of water bodies (rivers, lakes, transitional and near-shore coastal waters) by 2015. The WFD was followed up by the adoption in 2008 of the Marine Strategy Directive (MFD, 2008/56/EC, 2008), which extended to all European seas the need to achieve or maintain good environmental status in the marine environment by the year 2020.

In this context, this paper explores how the respective changes that have occurred in the last 24 years (1984–2007) in urban wastewater treatment and in agriculture have modified the quality of surface river waters in the Seine, Somme and Scheldt drainage basins, their nutrient delivery to the coastal zone, and the related coastal eutrophication visible in spring as undesirable blooms of *Phaeocystis* colonies (Lancelot et al., 1987; Rousseau et al., 1994). Previous papers have focused on the same studied zone computing N, P and Si budgets at the watershed scales (Thieu et al., 2009) but for only two or three contrasted hydrological years. Other papers computed fluxes delivered to the coastal zone over a long period on the Seine watershed (Billen et al., 2001, 2007b) or on the Scheldt one (Billen et al., 2005), with a non spatially distributed version of the Riverstrahler model. Some other papers were devoted to the marine ecosystem and their algal developments (MIRO model: (Gypens et al., 2007; Lacroix et al., 2007a, 2007b). Coupling between the Riverstrahler-model and a marine model Siam 3D was tested on the Seine Bight (Cugier et al., 2005) whereas the coupling of the models Riverstrahler and MIRO was explored over a long period for determining the riverine deliveries at the Belgian coastal zone. The GIS-based spatially distributed version of the Riverstrahler model (Seneque–Riverstrahler, SR, (Ruelland et al., 2007)) and its coupling with MIRO model were made possible for one year (2000, Lancelot et al., 2011) where some measures dealing with the improvement of WWTP or a better management of diffuse pollutions were explored in Thieu et al. (2010). The present paper goes further by modelling nutrients and riverine phytoplankton over 24 years on the Seine, Somme and Scheldt watersheds using this new, improved version of the SR model. The spatial distribution allows us to model the changes which occurred within the hydrological networks in terms of anthropogenic N and P concentrations from 1984 to 2007.

Compared to similar previous coupled river–coastal sea models applied in either the Seine–Eastern Channel and the Scheldt/Seine–Southern North Sea, the new spatially explicit application is well designed to calculate the seasonal and spatial variations of water quality within drainage networks ranging in scale from 10 to over 100,000 km^2. In addition, the method for calculating N diffuse sources released from agricultural practices was revised and now includes estimates of N surplus.

A complete year-to-year reconstruction of the changes in hydrology, agricultural practices and wastewater treatment over the 24-year period was carried out here, describing in detail the evolution of water quality within the drainage network of the three watersheds, mainly in terms of NO_3^- and PO_4^3, two major threats for freshwater water quality and drinking water production, in comparison with silica a key essentially diffuse nutrients. Further chaining this new version of the Seneque/Riverstrahler model implemented on the Seine, Somme and Scheldt, with the marine ecological MIRO model (Lancelot et al., 2005), made it possible to quantify the relationship between the changes in human activity in the watersheds and algal development in the adjacent coastal zone over the entire 24-year period. Particular attention is paid to the response of *Phaeocystis* bloom magnitude and duration to the quantitative and qualitative modifications of nutrient loads to the coastal sea.

2. Study site

The study focuses on the Seine, Somme and Scheldt (the "3S") watersheds located in the north of France, Belgium and The Netherlands (Fig. 1). Due to the dominant mean water circulation pattern (Turrell et al., 1992), nutrient loads cumulate along a SW–NE direction so that waters flowing in the Belgian coastal zone are directly impacted by the Scheldt and indirectly by the Seine and Somme rivers (Turrell et al., 1992). A total watershed area of 102,380 km^2 is drained by more than 26,300 km of rivers. The relief is smooth, less than 100 m in altitude on average, with a maximum elevation of 910 m in the southeastern part of the Seine basin (Table 1).

The geology is dominated by sedimentary formations. Limestone, clay and chalk formations of the Somme and Seine watersheds form concentric rings around Paris. Metamorphic formations are only present in the southern part of the Seine basin (Billen et al., 2007a). The Scheldt basin is dominated by loamy, sandy and loamy-sandy formations.

The climate is oceanic temperate with a yearly 630 mm of precipitation with a maximum during winter and a minimum during summer. The mean annual temperature is about 12 °C with a mean annual amplitude of 8 °C. Hydrology follows the climatic constraints of temperate regions, with minimum discharges observed at the end of summer and the maximum ones during winter (Fig. 2). Mean discharges for the 1984–2007 period are 527 m^3 s^{-1}, 140 m^3 s^{-1} and 35 m^3 s^{-1} for the Seine at Poses, Scheldt at Doel and Somme at Abbeville, respectively. The hydrology of the three watersheds is well synchronised, as can be observed from the moving average over 12 months (Fig. 2). Maximum discharges are observed in 1998, 1995 and 2001 for the three basins, while minimum ones are observed in 1992, 1996 and 2006. Some oscillations are evidenced with discharges increasing over 2 or 4 years and then decreasing over 1 or 4 years. Nevertheless, with a studied period of only 24 years, it is not possible to show any oscillatory phenomenon or a link with the North Atlantic Oscillations (NAO). Massei et al. (2010) showed that oscillations of NAO can explain 23% of total variance of Seine River flow. This kind of observations can be partially verified at the outlet of other watersheds of the region as that of the Rhine River (Ionita et al., 2012) or the Thames Rives (Marsh, 2001). It should be reminded however that the hydrology of the Seine and Scheldt basins is deeply impacted by human activities. Reservoirs have been built in the upstream part of the Seine basin in order to regulate discharges (Garnier et al., 1999) and some flow derivations are present within the Scheldt basin.

In 2006, arable land dominated within the three basins (Corine Land Cover 2006), at 77, 52 and 39% for the Somme, Seine and Scheldt, respectively. Arable lands are located in the centre of the basin of the Seine while for the Scheldt they are located in the southern part. Grasslands cover less than 10% of the three basins. The maximum is reached within the Seine basin with 9.8% of grasslands located on the edges of the basin, essentially in Morvan and in Normandy. Forests are scarce within the Somme and the Scheldt basins, i.e., 7% in each, but cover a quarter of the Seine basin, essentially at the northern, eastern and southern edges. Despite the weight of the Parisian agglomeration, urban areas only cover 7% of the Seine basin in the centre and along the main rivers. The Scheldt basin is more densely populated with some 25% of the basin devoted to urban areas homogeneously

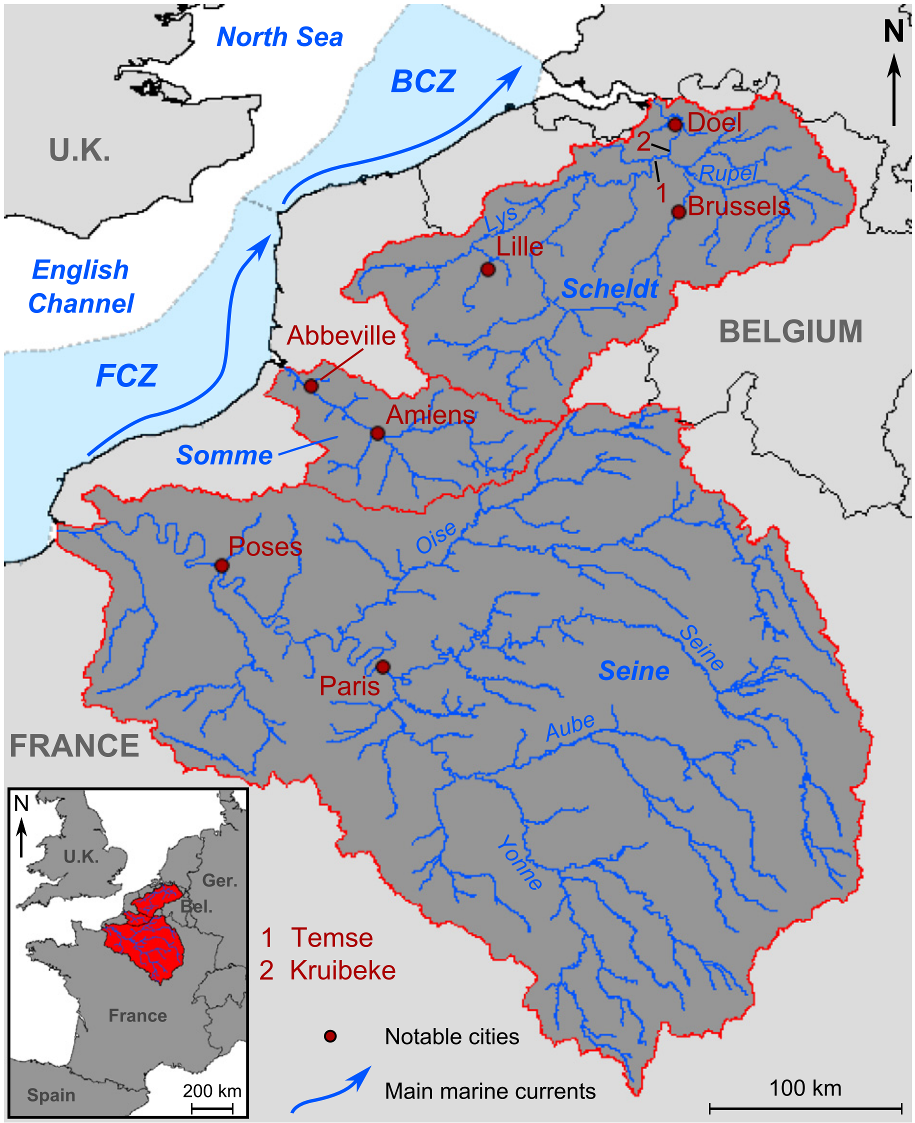

Fig. 1. Map of the domain studied locating the Seine, Somme and Scheldt watersheds and the main currents in the English Channel and the North Sea.

distributed within the watershed. The major physical and human characteristics of the three basins are summarised in Table 1.

3. Methods

3.1. The modelling chain, coupling the Seneque/Riverstrahler and MIRO models

The Seneque/Riverstrahler model (SR Model), a generic modelling tool coupled to a GIS-interface, was designed to calculate the seasonal and spatial variations of water quality within drainage networks ranging from 10 to over 100,000 km^2 (Ruelland et al., 2007; Thieu et al., 2009). The river network is represented as a combination of sub-basins and branches. Sub-basins are idealised by a regular scheme of confluence of tributaries with mean morphological characteristics by stream order (Strahler, 1957). These sub-basins are connected to branches represented with a higher, kilometric spatial resolution. This representation of the drainage network takes into account the biogeochemical processes occurring in both small first-order streams and large tributaries. The water flows in the hydrological network are calculated from the specific discharges generated within the watershed of the different sub-basins and branches considered, as calculated from recorded daily discharge at available gauging stations, and separated into surface runoff and base-flow components using the Eckhardt recursive filter (Eckhardt, 2008).

The principle of the SR model is to combine these water flows that are routed through the defined structure of basins and branches with a model describing the biological, microbiological and physical–chemical processes occurring in the planktonic and benthic realms of the water bodies. The module representing the kinetics of the biogeochemical processes is known as the Rive model and contains 30 state variables. These include nutrients (i.e. all inorganic, organic, dissolved and particulate N, P, Si forms), oxygen, suspended matter,

Table 1
Main characteristics of the Seine, Somme and Scheldt watersheds.

	Seine	Somme	Scheldt
Area (km^2)	76,270	6190	19,900
Maximum altitude (m)	910	223	217
Mean altitude (m)	97	55	26
Drainage density (km·km^{-2})	0.29	0.11	0.26
Mean discharge (m^3·s^{-1})	527	35	140
Population 2006	16,440,175	683,675	10,854,432
Population 1990	15,382,830	663,978	10,333,356

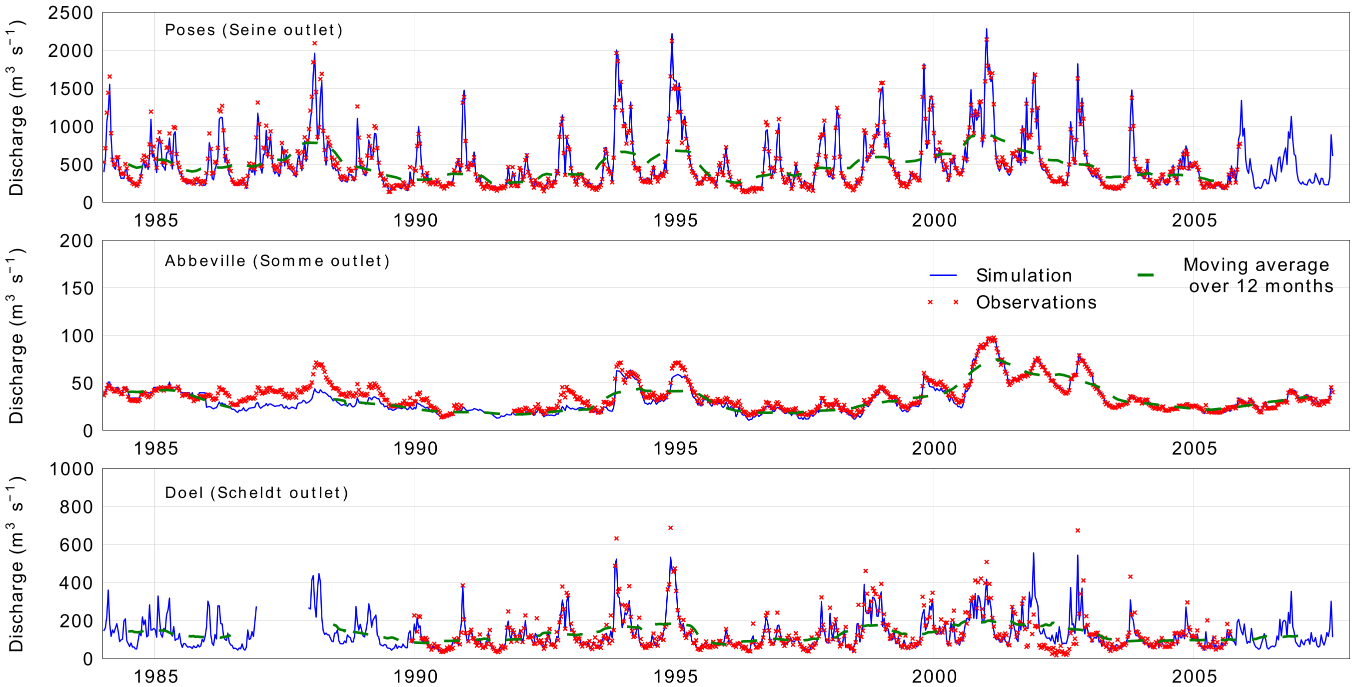

Fig. 2. Discharges at the outlet of the Seine (Poses), the Somme (Abbeville)and the Scheldt (Doel) Rivers for 1984–2007 (no data available for the year 1987 on the Scheldt). Red dots are measurements and blue lines are the SR Model simulations.

dissolved and particulate detrital organic carbon, and phytoplankton as diatoms and non-siliceous algae, bacteria and zooplankton (Garnier et al., 2002). Most processes important to the transformation, elimination and/or immobilisation of nutrients during their transfer within the river network are explicitly calculated, including algal primary production, aerobic and anaerobic organic matter degradation by planktonic as well as benthic bacteria with coupled oxidant consumption, nutrient remineralisation, nitrification and denitrification, and phosphate reversible adsorption onto suspended matter and subsequent sedimentation. The calculation scheme is based on a Lagrangian approach in which water bodies from each headspring are followed along the drainage network structure assuming a steady flow regime. The seasonal variations are described by considering successive steady flows every 10 days over the whole year cycle. A detailed description of the Rive Model equations and parameters is reported in Garnier et al. (2002). Besides morphological and climatic constraints, the SR Model considers diffuse and point sources of nutrients from land-based anthropogenic sources (see below).

SR outputs at the river outlets allow one to quantify nutrient fluxes delivered to the sea as well as the ICEP index, an indicator of the eutrophication potential (Billen and Garnier 2007; Garnier et al., 2010).

MIRO is also a mechanistic biogeochemical model and was designed to asses and understand eutrophication problems associated with *Phaeocystis* blooms in coastal zones (Lancelot et al., 2005, 2007, 2011). Similar to the RIVE model, MIRO includes 38 state variables assembled in four modules describing the dynamics of phytoplankton (diatoms, nanoflagellates and *Phaeocystis*), zooplankton (copepods and microzooplankton), the degradation of dissolved and particulate organic matter (each with two classes of biodegradability) and the regeneration of inorganic nutrients (NO_3^-, NH_4^+, PO_4^{3-} and $Si(OH)_4$) by bacteria in the water column and the sediment. Equations and parameters were formulated based on current knowledge of the kinetics and the factors controlling the main auto- and heterotrophic processes involved in the functioning of the coastal marine ecosystem (fully documented by Lancelot et al. (2005) and in http://www.int-res.com/journals/suppl/appendix_lancelot.pdf). For this application, MIRO is implemented in a multi-box system, arranged in successive boxes from the Seine Bight to the Belgian Coastal Zone. Each box receives water from the upstream adjacent box and adjacent rivers and exports water to the downstream adjacent box. The boxes and geographical features are described in Lancelot et al. (2005) and the residence times are calculated based on results of the COHERENS-3D hydrodynamical model (Lacroix et al., 2004), as described in Gypens et al. (2007).

The similar structure of the SR and MIRO models makes it feasible to combine them (Lancelot et al., 2007), so as to calculate the effect of land-based sources of nutrients derived from human activity in the watersheds on coastal eutrophication.

4. Input data to the SR model

SR–MIRO simulations were conducted over the 1984 period making use of meteorological conditions and point and diffuse sources of nutrients as constraints.

4.1. Point sources

Point sources are industrial and urban wastewater released into the river network via (or not) wastewater treatment plants (WWTPs). Each WWTP is characterised by a number of treated equivalent inhabitants and the type of treatment applied (namely primary, secondary or tertiary treatment, including P removal, nitrification and denitrification). For the years after 2000, these data were obtained from Water/Environmental Agencies in France and the Walloon and Flemish regions for Belgium. For the 1980s and 1990s, the point sources were reconstructed taking into account population dynamics and the rate of connection to sewer systems within each watershed, the year of the implementation of each WWTP and assumptions on the future improvements in wastewater treatment. Within the Scheldt and the Somme watersheds, industrial releases are assumed to be mainly collected by urban sewers and to reach the rivers via domestic WWTPs. Within the Seine watershed, industrial discharges are treated within their own wastewater treatment.

4.1.1. Population changes within the 3S basins

The French "Institut National de la Statistique et des Sciences Economiques" (INSEE) and the Belgian Federal Government released population census figures for the years 1990 and 2006. In 2006 the total population of the 3S was about 28 million inhabitants (INSEE/ INS), compared to 26.4 in 1990, i.e. an overall increase of 5.7%. The population is not uniformly distributed within the 3S watersheds. In 2006, the Scheldt basin was the most densely populated with 540 inh km^{-2}. The highest densities are located in the central part of the basin around Brussels and in the northern part around Antwerp. The density within the Seine basin is 215 inh km^{-2} on average, but presents a significant disparity. Within the Parisian agglomeration, the fourth in Europe with roughly 12 million inhabitants, the density can reach more than 5000 inh km^{-2}, while large rural areas have fewer than 20 inh km^{-2} in the upper parts of the basin. The Somme watershed is less populated and the average density is 110 inh km^{-2}.

Between 1990 and 2006, the population increased in the three watersheds, by 6.9%, 3.0% and 5.0% for the Seine, Somme and Scheldt basins, respectively. This increase was not uniformly distributed: the density decreased in the upper part of the Seine watershed and increased in the Parisian agglomeration and in the downstream part of the watershed. Population density also increased in the Northern part of the Scheldt basin.

4.1.2. Improvement of wastewater collection and treatment: the success story of P abatement

In the middle of the 1980s, the reduction of urban and industrial point sources of organic matter, phosphorus (P) and ammonium (NH_4^+) pollution was the main challenge for river water quality. In-stream organic matter degradation and nitrification of NH_4^+ released by insufficient WWTPs were indeed leading to severe oxygen depletion in large sectors of the drainage network, especially the Seine downstream from Paris (Billen et al., 2007b) and the Zenne and Rupel downstream from Brussels (Billen et al., 1985; Garnier et al., 2013 this issue). Regarding P, the specific discharge was about 4 mgP l^{-1} day^{-1} inh^{-1} and was not or poorly treated within the WWTPs, so that P was largely in excess in comparison to both Si and N in downstream river sectors (Billen et al., 2001, 2005).

In early 1990, the situation improved for two main reasons. The first one was the banishment of polyphosphates from washing powders, which rapidly reduced the specific P discharge to about to 2 mgP l^{-1} day^{-1} inh^{-1}. This banishment occurred earlier in Belgium, during the late 1980s and the early 1990s, than in France where it occurred during the mid- and late 1990s due to the resistance of the French industrial lobbies (Billen and Garnier, 1999). The specific P discharge is nowadays about 1 mgP l^{-1} day^{-1} inh^{-1}, close to the value of physiological release (Verbanck et al., 1994). The second reason was the gradual implementation of P treatment in WWTPs (Even et al., 2007). In compliance with the urban wastewater treatment directive (91/271/CEE, 1991b), municipalities with more than 2000 inhabitants are obliged to treat their wastewaters before their release into rivers. Cities located within NO_3^--sensitive areas have to implement a tertiary treatment in order to decrease the P discharge by about 85% and the N discharge by about 70%. The largest fraction of Parisian treated wastewaters is released into the Seine at Achères, some 50 km downstream from Paris. This WWTP is one of the largest in Europe, treating more than 1,700,000 m^3 day^{-1}of wastewater, and was equipped with a P tertiary treatment in 2000; it was upgraded to nitrify all NH_4^+ beginning in 2007. A 70% denitrification will be achieved in 2015.

Within the Scheldt watershed, until the beginning of the 21st century Brussels wastewater was not treated at all and was directly released into the Zenne River (Garnier et al., 2013 this issue). In 2000, a first WWTP, "Brussels South", treating about 360,000 equivalent inhabitants, with a rather basic activated sludge process, was implemented a few kilometres upstream from Brussels and was fully operating in 2001. A second one, "Brussels North", treating both P and N of some 1,100,000 equivalent inhabitants, was implemented a few kilometres downstream from Brussels in 2007.

The reconstruction of N and P deliveries of WWTPs over the period (Fig. 3) shows that the release of P in the Seine quickly decreased from 1988 to 1994 and up to 2007 (Fig. 3 a), while for the Scheldt a 50% decrease is observed in 1989–1990 (Fig. 3 b). N domestic inputs slightly increased in the middle of the 1990s in the Seine, then decreased slightly and quickly in 2007 with the implementation of tertiary treatment (Fig. 3 a). On the Scheldt basin, N gradually increased from 1984 to 2003, stabilised for some years before it quickly decreased in 2006 and 2007 (Fig. 3 a) after the implementation of Brussels North WWTP.

4.2. Diffuse sources

Lithology and soil properties, land use, agricultural practices and climatic constraints together determine the diffuse sources of nutrients (N, P, Si) to the hydrosystem.

Diffuse sources are determined based on land use (Corine Land Cover 2006) and agricultural practices (official French and Belgian agricultural statistics).

Twelve major agricultural regions have been distinguished within the 3S watersheds: these are the Belgian agricultural zones (Campine, sandy region, sand-loamy region, loamy region) for the Scheldt, and groupings of French Small Agricultural Regions as defined by Mignolet et al. (2007) for the Seine and Somme watersheds (Fig. 4). For each land use type and each of the 12 agricultural regions, a mean constant concentration is provided for sub-root water and groundwater concentrations, respectively, which are derived as described below.

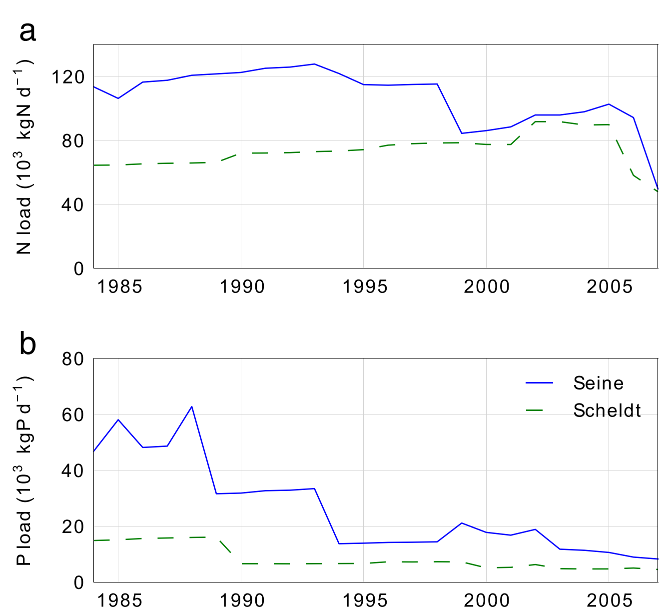

Fig. 3. Reconstruction of the total N (a) and P (b) loading from domestic wastewater facilities into the Seine and Scheldt drainage network. Data were communicated by Water Agencies from 1999 to 2007 for the Seine and for 2006 and 2007 for the Scheldt, but estimated from information on the population, sewer connection data and WWTP implementation dates for earlier periods.

The effect of agriculture on water quality is far from being direct, however, as a significant fraction of the released nutrient finds its way through aquifers with residence time on the order of several decades in the studied area.

4.2.1. Minor land use changes

Changes in land use between 1990 and 2006, as described by the Corine Land Use data base, are minor. The most important trends are a 1.8% increase in urban areas within the Scheldt basin, and a 0.60%, 0.43% and 0.42% decrease in areas devoted to grasslands for the Seine, Somme and Scheldt basins, respectively. Other land use classes are quite stable spatially. Changes were greater for agricultural practices than land use.

4.2.2. Changes in the N balance of agriculture

The long-term trajectory of agricultural development during the last 50 years greatly differs between the different agricultural regions of the three basins. The regions of the central part of the Seine basin have gradually specialised in cereal and industrial crops and are characterised by very low livestock densities (<0.3 LU ha^{-1} of agricultural land, where livestock unit (LU) represents the amount of livestock equivalent to a modern milking cow and excreting 85 kgN yr^{-1}). By contrast, very intensive livestock farming (>5 LU ha^{-1}) has increased in the northern part of the Scheldt basin, whereas livestock densities were maintained or even lowered in the peripheral regions of the Seine basin, e.g. in the Morvan region (Fig. 5 a). The regions also differ in their use of synthetic fertilisers (Fig. 5 b). While already common in Belgium in the 1950s, the use of industrial N fertilisers increased only in the 1960s and the 1970s in most French areas, where it reached 230 kgN ha^{-1} yr^{-1} in the Brie–Beauce and Champagne regions, i.e. close to the current levels in Belgium. In recent years, the rate of synthetic N fertilisation application has stabilised or decreased in all other regions, as for P fertilisers. In order to characterise the past trajectory of each agricultural region since the 1950s in terms of N use, a complete N budget of arable land was calculated from agricultural statistics available at some key periods: urbanisation since the 1950s, industrialisation of agriculture since the 1970s and the European reforms since the 1990s. Total N output by exported crops was calculated from production figures and converted into N content using standard coefficients (Billen et al., 2009). Total fertilisation was calculated as the sum of synthetic fertiliser and manure application, biological atmospheric N fixation by legumes, and atmospheric deposition. The details of the calculations and hypotheses are provided in the supplementary material section. For each region, we then plotted the average N crop export per hectare cropland against total N fertilisation (Fig. 6). The data were fitted by a curve with the following equation:

$$\text{Nexport} = \text{Ymax} \times [1 - \exp(-\text{fertilisation}/\text{Ymax})] \quad (1)$$

where Ymax represents the maximum yield at saturating fertilisation.

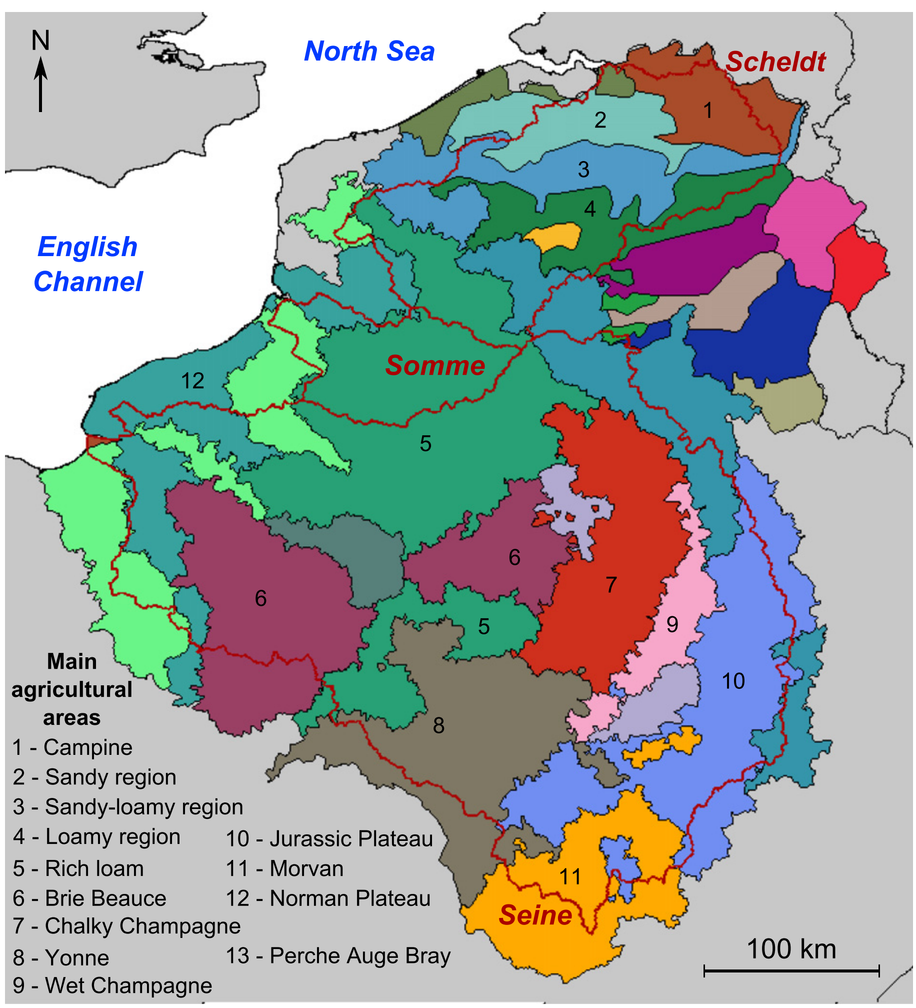

Fig. 4. Agricultural areas within the Seine, Somme and Scheldt watersheds as taken into account in the modelling approach for determination of diffuse sources, together with surplus (see text for explanation).

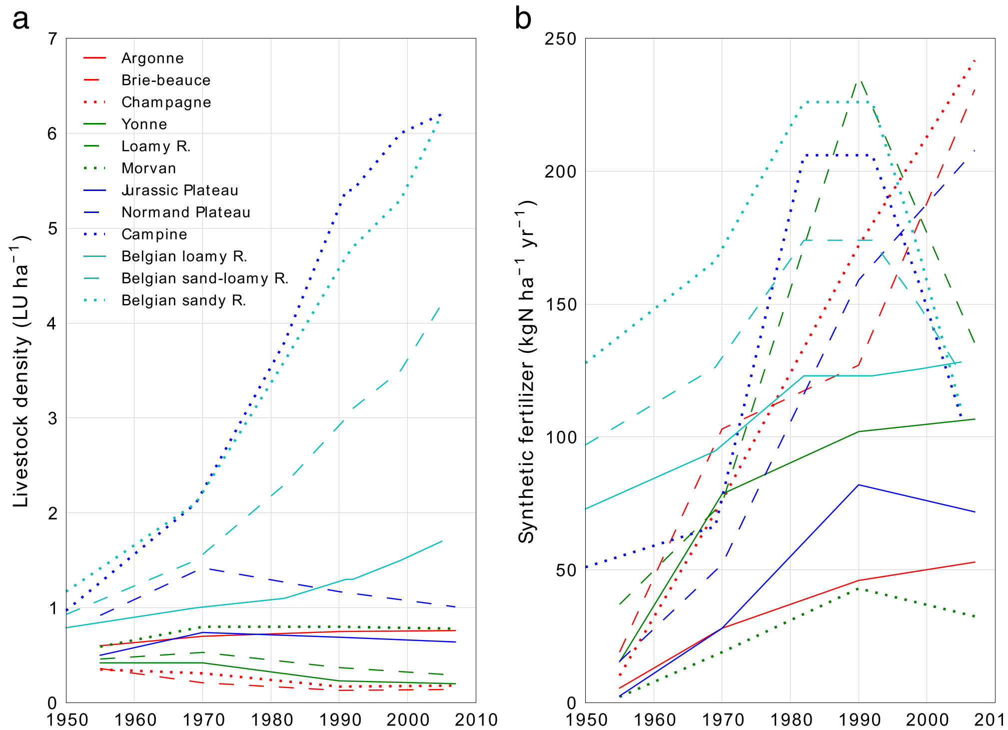

Fig. 5. (a) Historical changes in livestock density (expressed as LU ha^{-1} of cultivated area) in the main agricultural regions of the Seine and Scheldt basin during from 1950 to today. (b) Historical changes in the rate of synthetic nitrogen fertiliser application (expressed as kgN ha^{-1} of cultivated area and per year) in the main agricultural regions of the Seine and Scheldt basins from 1950 to the present. Sources: National Institute of Statistics (INS) data by agricultural region for Belgium; Agreste for France, data for one or two departments are used as representative for the whole agricultural area to which they belong.

For most French agricultural regions, the trend obtained shows a general increase of total fertilisation over the 1950–2005 period, accompanied by a much more limited increase of production, thus a gradual decrease of the N use efficiency (Fig. 6 a, b). The same conclusion holds for the agricultural regions of the Scheldt basin until the 1990s, with even higher fertilisation rates and lower efficiency rates.

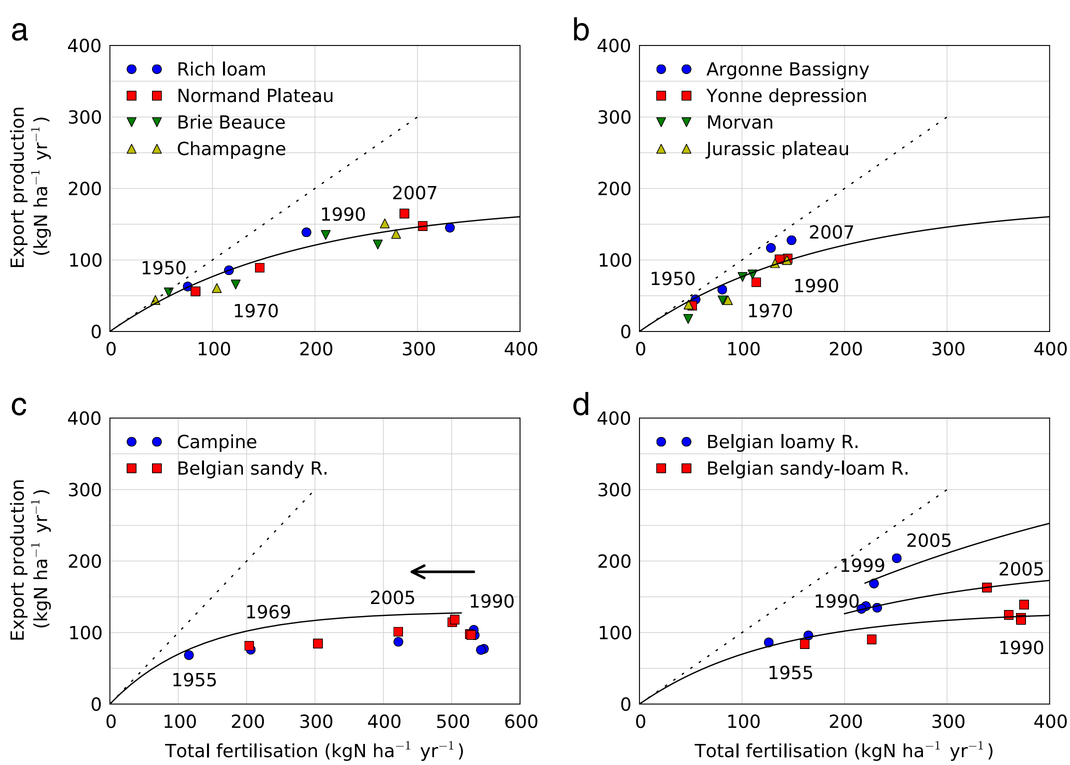

Fig. 6. N crop export vs. total N fertilisation of arable land in the different agricultural areas of the Seine, Somme and Scheldt basins from 1950 to 2005. A fitted curve with equation Nexport = Ymax × [1 − exp(−fert / Ymax)] is also indicated for each data set.

However, changing trends have become apparent in the past few decades. In the Southern regions (Fig. 6 d), an increase in Ymax can be observed, suggesting better N use efficiency and fewer losses. In the Northern regions (Fig. 6 c), the total fertilisation rate, although still quite high, has been decreasing since the 1990s. This last trend corresponds to the implementation of public policies aiming at reducing agricultural N pollution. While these policies are still only incentive in France, they were compulsory in the Walloon region of Belgium, where a public organism (Nitrawal) is in charge of systematic control through cropland soil analysis, and in the Flemish region, where treatment of livestock manure and export of N residue have been strongly encouraged by public financial support. In 2008, the Flemish region has treated and exported, respectively, 20% and 30% of the N and P content of its total livestock waste production.

4.2.3. Diffuse source assessment

The difference between N crop export and N fertilisation (the vertical distance between each point and the diagonal line in the diagrams of Fig. 6) represents the N-surplus, i.e. the excess fertilisation over the N taken up by crops. This surplus is available for leaching, gaseous emission or storage as organic N in the soil.

The N concentration of surface runoff characterising the different land use classes and agricultural areas of the three basins (Table 2) was assessed based on the assumption that, for well-drained arable soils, leaching is by far the main fate of the agricultural surplus; when divided by the runoff depth, it provides a good estimate of NO_3^- concentration of sub-root water (Table 2). This approach cannot be used, however, for hydromorphic soils, like those often found in Northern Belgium, where denitrification as N_2 gas is the major fate of the N surplus. Nor can it be used for permanent grassland soils, which accumulate very large amounts of N in the soil organic pool and are subject to rather limited N leaching compared to arable land. The base-flow concentration was estimated from previous studies (Curie et al., 2011; De Becker et al., 1985; Fritz, 1994; Poitevin, 1997; Roberts and Marsh, 1987; Rousseau et al., 1986; Strebel et al., 1989; Vandenberghe, 2010) on groundwater concentration available for the major aquifers of our study area (Fig. 7); in regions without large aquifers, the same concentration was taken for surface and base-flow components.

In the SR model, these values are used to characterise the diffuse sources of nutrients.

5. Results

5.1. Validation of the SR model

Fig. 8 compares SR simulations of water quality variables (nutrients, phytoplankton) at Poses (the outlet of the basin, upstream from the estuarine zone) with observations for the 1985–2007 period, provided by the Seine Normandy Water Agency (Agence de l'Eau Seine Normandie, Réseau National de Bassin). The discharge during the period varied greatly, with the driest years in 1996 and 2005 and the wettest in 2001 and 1995 (Fig. 2). We calculated three indicators of the goodness of fit of the RS simulations. The RMSE (root mean square $error = \sqrt{(\sum(vobs-vcalc)^2)}$ is a measure of the inaccuracy of the model's prediction. The bias $\left(\left(\sum\frac{|vobs-vcalc|}{vobs}\right)/n\right)$ is a measure of the systematic relative over- or underestimation by the model. The Bravais–Pearson R $\left(r=\frac{covar(x,y)}{\sqrt{var(x)\times var(y)}}\right)$ measures how well a model fits observations (Allen et al., 2007; Krause et al., 2005). Table 3 gathers the values of these estimators for the most important variables. Collectively, these indicators show a generally good fit between calculation and observations for most variables, especially for P, with a highly significant Bravais–Pearson R higher than 0.5. The goodness of fit is lower for NO_3^- and NH_4^+; this is particularly true for the 1980s and early 1990s, probably in part because of the difficulty of obtaining reliable data on point sources of NO_3^- and NH_4^+ at that time. For phytoplankton, despite a 62% bias, the seasonal variations are well captured.

For the Scheldt River, simulations were extracted at Kruibeke for the 1996–2007 period and at Temse for the 1984–1995 period and were compared with available data provided by the Vlaamse Milieumaatschappij (VMM), l'Unité de Gestion du Modèle Mathématique de la Mer du Nord (UGMM) and the Administration Sea and Watercourses (AWZ) (Fig. 9). The driest years were 1990 and 1996 and the wettest ones were 2001 and 1988 (Fig. 2). Regarding water quality, whereas simulations did not fit the observations as well at Kruibeke and Temse as at Poses using RMSE indicator, taking into account the Bravais-Pearson R, the agreement is reasonable. Besides the less numerous data for validation, the lower model performance between the two watersheds can be explained by the forcing data that were more difficult to reconstruct over the 24-years period for the Scheldt watershed. The performance of the SR model applied to the Somme could not be assessed due to insufficient observations available.

5.2. Interannual variations in water quality at the river outlet

5.2.1. Seine River

The long-term variation of NO_3^- concentrations at the outlet of the Seine River brought out two distinct periods (Fig. 8). The first one, from 1984 to 1998, showed a slight NO_3^- annual increase, from 4.5 to 6.7 mgN–NO_3^- l^{-1}. The period after 1998 was characterised by concentrations stabilising around 6.7 mgN–NO_3^- l^{-1}. Over the period, the slope of the simulated concentration trend line in function of time shows an increase of 44.70%. The seasonal variations follow the hydrological regime with lower NO_3^- concentrations by low discharge. Overall, the interannual variations of NH_4^+ (Fig. 8) concentrations mainly

Table 2
Sub-root and phreatic water nitrate concentration (in mgN L^{-1}) for the arable land of the different agricultural regions considered in the watershed area of the Seine, Somme and Scheldt (grdwater = groundwater).

Nitrate (mgN l^{-1})	Runoff (mm yr^{-1})	1960 Sub-root	grdwater	1970 Sub-root	grdwater	1980 Sub-root	grdwater	1990 Sub-root	grdwater	2000 Sub-root	grdwater	2010 Sub-root	grdwater
Rich loam	243	8.0	6.0	25.0	7.0	33.0	9.0	41.0	12.0	57.0	12.5	62.0	13.0
Brie Beauce	197	10.0	6.0	18.0	7.0	30.0	9.0	41.0	12.0	56.0	12.5	56.0	13.0
Champagne	278	7.0	2.0	13.0	4.0	22.0	6.0	29.0	7.0	40.0	8.0	47.0	9.0
Normand Plateau	229	9.0	6.0	26.0	7.0	35.0	9.0	44.0	12.0	61.0	12.5	66.0	13.0
Yonne depression	246	6.1	6.1	6.1	4.1	5.1	5.1	5.5	5.5	6.1	6.1	8.1	8.1
Morvan	327	3.1	3.1	3.1	3.1	3.8	3.8	4.1	4.1	4.6	4.6	6.1	6.1
Argonne Bassigny	295	3.4	3.4	3.4	3.4	4.2	4.2	4.6	4.6	5.1	5.1	6.8	6.8
Jurassic Plateau	341	2.9	2.9	2.9	2.9	3.7	3.7	4.0	4.0	4.4	4.4	5.9	5.9
Belgian loamy R.	234	12.8	4.0	21.4	5.0	25.7	6.0	23.6	7.0	23.6	8.0	19.3	9.0
Belgian sand-loamy R.	273	14.6	2.0	21.9	4.0	36.6	7.0	54.9	9.0	54.9	11.0	43.9	12.0
Belgian sandy R	240	16.7	16.7	41.7	41.7	41.7	41.7	50.0	50.0	33.4	33.4	29.2	29.2
Campine	351	14.3	14.3	28.5	28.5	28.5	28.5	34.2	34.2	22.8	22.8	20.0	20.0

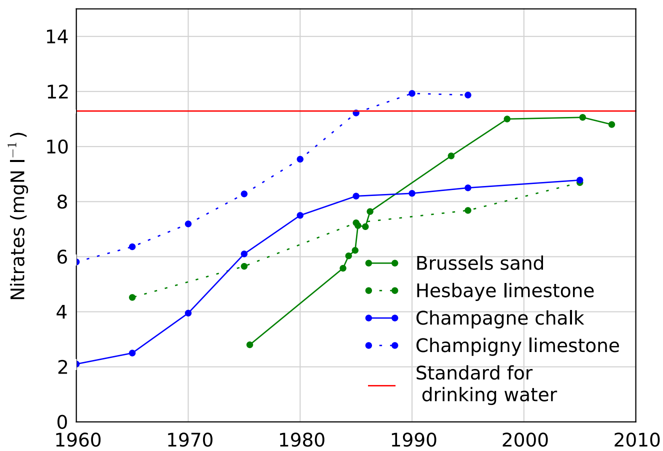

Fig. 7. Nitrate concentration in the major aquifers of the Seine, Somme and Scheldt basins.

reflect the gradual improvement of wastewater treatment as well as a trend related to the hydrological regime, with higher values at high discharge. From 1984 to the first half of the 1990s, the maximum concentrations of NH_4^+ reached 5 or 6 mgN–NH_4^+ l^{-1}, while they have not exceeded 3 or 4 mgN–NH_4^+ l^{-1} since the year 2000. From 1987 to 2007, a general decrease of 51%. The spectacular decrease in P concentrations observed (Fig. 8) was well captured by the SR model, closely related to the improvement of wastewater treatment plants. Seasonal variations again follow the hydrological regime, with higher concentrations by low discharge. In the second half of the 1980s, the maximum concentrations of phosphates reached about 2 mgP l^{-1} with a yearly average around 1 mgP l^{-1}. Since 2000, the maximum concentrations have been approximately 0.4 mgP l^{-1} and the yearly average concentration less than 0.2 mgP l^{-1}. The percentage of decrease is the highest with a value of 90% from 1984 to 2007. Si follows a clear regular seasonal pattern with a winter maximum of 10 mgSiO$_2$ l^{-1} and a spring/summer depletion explained by diatom growth (Fig. 8). No clear trend is observed and the slope of the simulated Si trend line is insignificant, showing a decrease of 5% over the period. As shown by Sferratore et al. (2006), human activities had little impact on the Si cycle, and the interannual variability was dictated by the succession of dry and wet years, which favoured or prevented algal development. Phytoplankton seasonal fluctuations mirrored the Si variations (Fig. 8), indicating that diatoms dominate the freshwater phytoplankton community. Interestingly, the spring maximum reached by phytoplankton showed significant interannual variations (40–80 μg Chla l^{-1}; Fig. 8) with no regular trend related to the long-term P decrease, suggesting that no limitation by this element has yet been reached, at least in the downstream Seine drainage network. Summer phytoplankton development at Poses, of lesser amplitude and generally dominated by green algae (Garnier et al., 1995; and unpublished data) are not well represented by the model. As green algae are very rapidly mineralised once in the salinity gradient, the simulations might not affect per se the total nutrient load delivered to the sea.

5.2.2. Scheldt River

Unlike in the Seine, simulated and observed NO_3^- concentrations at the Scheldt outlet were rather stable during the simulated period, i.e. varying between 4 and 6 mgN–NO_3^- l^{-1} (Fig. 9). This is indicated by the lower value of the slope of the trend line of simulated NO_3^- in Kruibeke showing a slight decrease of 8% over the period. From 1984 to the end of 1990, the simulated maximum concentrations reached 8 mgN–NH_4^+ l^{-1} with a yearly average at 4.1 mgN–NH_4^+ l^{-1}. The simulated trend of NH_4^+ (Fig. 9) showed a marked decrease after 1997, most probably in response to gradual improvement of wastewater treatment in the watershed. After 2000, the maximum simulated concentrations reached 4 and 5 mgN–NH_4^+ l^{-1} in Temse and Kruibeke, respectively, and were slightly overestimated compared to observations. Overall the decease of NH_4^+, from 1984 to 2007, is 30%. In 2007, thanks to the implementation of the new Brussels North WWTP, the concentration

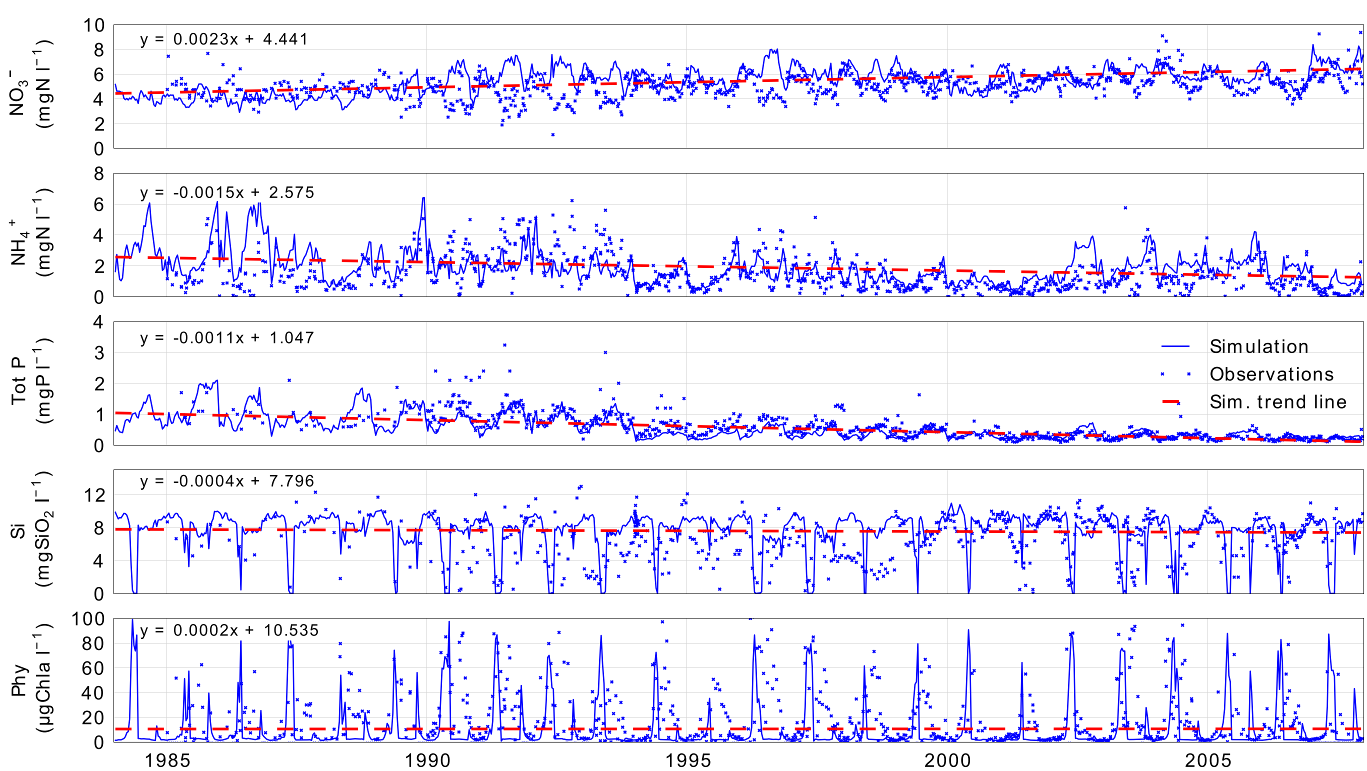

Fig. 8. Simulation of NO_3^-, NH_4^+, total P, Si, and phytoplankton compared to observations at Poses (Seine outlet) for 1984–2007.

Table 3
RMSE, bias and Bravais-Pearson R for the SR Model simulations at Poses, Kruibeke and Temse (** significance between 0.001 and 0.01, * significance between 0.02 and 0.1).

	Phyto	SM	NO_3^-	NH_4^+	PO_4^{3-}	P tot	SiO_2	O_2
Seine: Poses								
RMSE	1.52	1.39	0.06	0.04	0.01	0.01	0.15	0.10
Bias	62.43	−36.41	−8.55	−22.85	−10.90	18.52	8.16	0.92
R	0.24 **	0.57 **	0.09 *	0.50 **	0.74 **	0.63 **	0.38 **	0.58 **
Scheldt: Kruibeke								
RMSE	7.04	8.43	0.23	0.42	0.02	0.05	0.49	0.56
Bias	27.13	−15.70	−26.23	−121.22	−55.26	13.06	−8.45	−119.73
R	0.35 *	0.27 *	−0.20	0.35 *	0.63 **	0.08	0.73	0.65 **
Scheldt:Temse								
RMSE			0.59	0.50	0.091	0.21		0.85
Bias			−58.05	20.70	−1.26	35.19		−92.72
R			0.55 *	0.60 *	0.81 **	0.16		0.62 **

was less than 1 mgN–NH_4^+ l^{-1}, still overestimated compared to observations.

The simulated P concentration (Fig. 9) was maximum between 1984 and 1987, amounting to 3 mgP l^{-1} (yearly mean, 1.8 mgP l^{-1}) but began a sharp decrease in 1989 responding to the P banishment in washing powders, slightly earlier than for the Seine (see also Billen et al., 1999). A second decrease is visible after 1998, corresponding to the implementation of P treatment in WWTPs (Fig. 9). Note that the purification of the agglomeration of Brussels was efficiently operating in 2007. The maximum concentrations reached in 2000 were about 1 mgP l^{-1} and the yearly mean was approximately 0.5 mgP l^{-1}. Consequently, the P decrease at Kruibebeke reaches 70%.

The simulated interannual variations of Si and phytoplankton were in better agreement with the observation that those obtained in the Seine River. The minimum Si concentration occurred in summer at the time of maximal phytoplankton growth, and maximum values reached in winter (13 mgSiO_2 l^{-1}). The maximum level of phytoplankton was observed in early summer; concentrations reached 60–100 μgChla l^{-1} depending on the year as reported by (Kromkamp and Van Engeland, 2010).

5.2.3. Somme River

Concerning the interannual simulation for the Somme basin discharges ranged from 12 to 105 m^3 s^{-1}, respectively, in 1996 and 2001, with a yearly average of 36 m^3 s^{-1} (Fig. 2). Concentrations are not shown due to the lack of validation data over this long term period, see Table 4 for changes in simulated fluxes. However, we can mention a mean annual NO_3^- concentration increased from 5 to 7 mgN–NO_3^- l^{-1} over the simulated period due to the large part of the basin devoted to agriculture. Total P followed the same trend as in the Seine and Scheldt watersheds and decreased from 0.3 to 0.08 mgP l^{-1} over the simulated period. Si and phytoplankton were quite stable over the period studied, 14 mgSiO_2 l^{-1} and 7 μgChla l^{-1} with some peaks at 30 μgChla l^{-1} in the late 1990s.

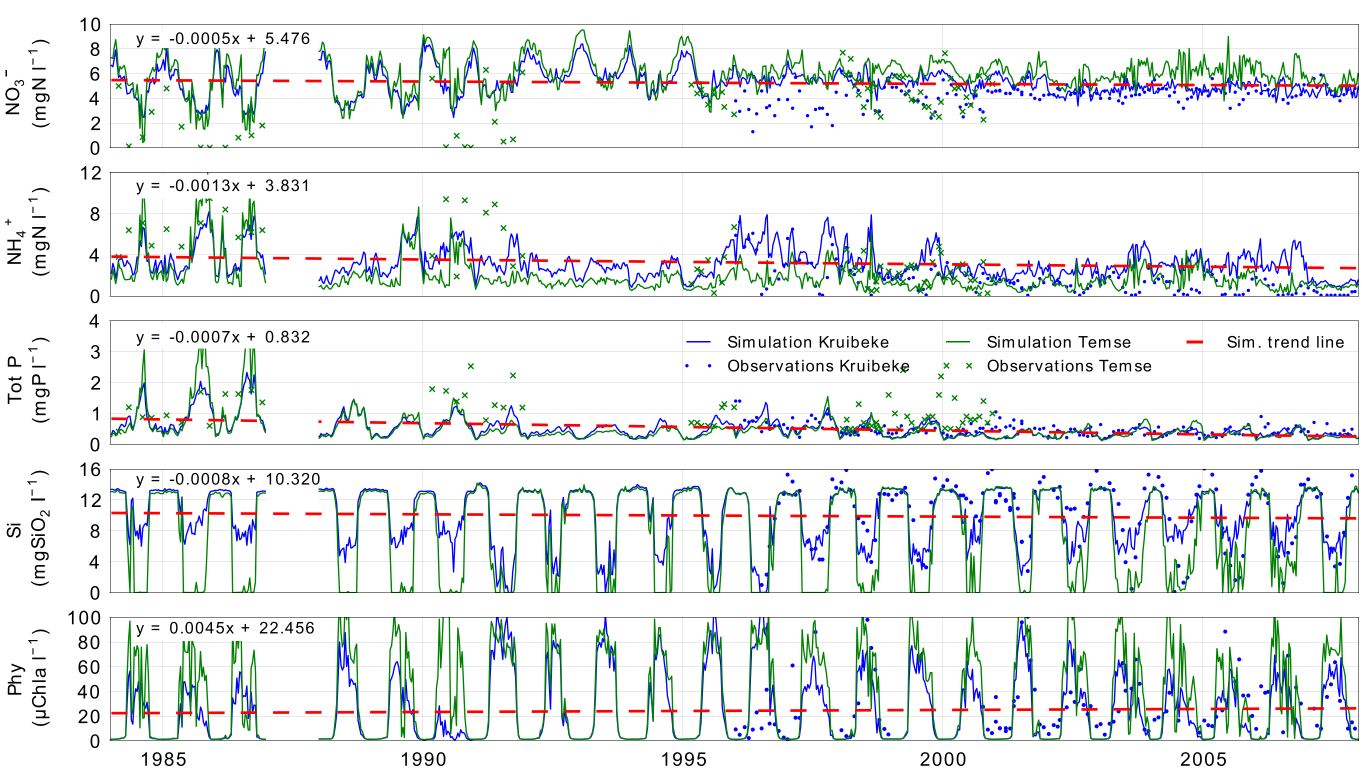

Fig. 9. Simulation of NO_3^-, NH_4^+, total P, Si, and phytoplankton (compared to observations at Kruibeke and Temse (Scheldt outlet) for 1984–2007 (no hydrological constraints available for the year 2007).

5.3. Water quality in the upstream river network

The SR model provides a comprehensive simulation of water quality at any point of the drainage network of the 3S rivers, which is also of great importance for human activities (Vörösmarty et al., 2010). Fig. 10 provides a summarising view of these results as maps of average NO_3^- and phosphate concentrations in all tributaries. P concentrations within rivers strongly decreased between 1985 and 2007 (Fig. 10 a, b). In 1985, ortho-phosphate concentrations in the low-order rivers of the upper parts of the basins were less than 0.16 mgP l^{-1}. PO_4^{3-} concentrations in higher-order rivers, such as the Marne, Oise and Somme, ranged from 0.16 to 0.64 mgP l^{-1}. In the downstream part of the Seine River, values higher than 0.64 mgP l^{-1} were reached. The situation within the Scheldt basin was the worst due to the higher population density and PO_4^{3-} concentrations reached more than 0.64 mgP l^{-1} in almost two-thirds of the drainage network (Fig. 10 a). The situation greatly improved in 2007 with PO_4^{3-} concentrations in most of the Seine and the whole Somme watershed less than 0.16 mgP l^{-1}. Only PO_4^{3-} concentrations in the part of the Seine River downstream of the Parisian agglomeration remain high, ranging from 0.16 to 0.32 mgP l^{-1}. In the Scheldt watershed, where PO_4^{3-} concentrations were less than 0.16 mgP l^{-1} upstream, but increase rapidly to 0.3 mgP l^{-1} or more in intermediate rivers (Fig. 10 b).

NO_3^- concentrations within the three watersheds did not follow the same trend. In 1985 NO_3^- concentrations in the upper parts of the Seine basin (Yonne, Aube, Marne) were less than 2.25 mgN–NO_3^- l^{-1}. In the major parts of the Seine and Scheldt basins, NO_3^- concentrations were less than 5.65 mgN–NO_3^- l^{-1}. Only in the Somme basin and in the French upper part of the Scheldt basin, did NO_3^- concentrations reach 11.3 mgN–NO_3^- l^{-1} (Fig. 10 c). In 2007, NO_3^- concentrations were higher everywhere within the three watersheds and exceeded the value of 2.25 mgN–NO_3^- l^{-1}. In the southern part of the Scheldt Basin, in the whole Somme basin and in several sub-basins of the Seine, NO_3^- now ranges from 5.65 to 11.3 mgN–NO_3^- l^{-1} (Fig. 10 d).

5.4. Nutrient fluxes to the coastal zone

Annual specific nutrient inputs to the coastal zone are estimated from N, P and Si provided by SR simulations at the outlet of the Seine, Somme and Scheldt rivers, respectively at Poses, Abbeville and Kruibeke for the simulated period (Figs. 8 and 9). Obviously, the most striking feature is the strong dependence of these nutrient fluxes on the concomitant interannual variation of discharge, which undergoes, over the period studied, a succession of dry and wet years with a 6- to 7-years periodicity. The reduction of nutrient sources when it occurred is superimposed and somehow masked by this hydrologic trend. Thus, Si fluxes, not significantly affected by human activities, are very much dependent on hydrological conditions. Si reached a maximum in the wettest year in 2001 for the Seine (2050 kgSi km^{-2}) (Fig. 11 a), the Scheldt (1735 kgSi km^{-2}) (Fig. 11 b) and the Somme (2800 kgSi km^{-2}), with annual averages of Si fluxes for the period 1984–2007 of 1005, 1130 and 1215 kgSi km^{-2} yr^{-1}, respectively. For the Seine at Poses, maximum N fluxes observed during the wettest years 2001 and 1995 are twice those of dry years, reaching 3175 kgN km^{-2} and 2760 kgN km^{-2}, respectively, versus 1415 kgN km^{-2} and 1440 kgN km^{-2} in 1989 and 1996 (Fig. 11 a). For the Scheldt at Kruibeke, maximum N fluxes similarly occurred during the wettest years in 2001 and 1988, reaching 2665 kgN km^{-2} and 2550 kgN km^{-2}, respectively, but the minimum N flux, 1390 kgN km^{-2}, was observed in 2007, not the driest year, reflecting in part the effect of implementation of the Brussels North wastewater treatment plant (Fig. 11 b). Regarding P inputs, a two-thirds decrease was observed at Poses from 290 kgP km^{-2} in 1985 to 90 kgP km^{-2} in 2007, i.e. 69% (Fig. 11 c; Table 4), by 49% at the outlet of the Scheldt, from 210 to 110 kgP km^{-2} (Fig. 11 d), and by 33% at the outlet of the Somme, from 85 to 55 kgP km^{-2} (see also Table 4).

The OSPAR convention recommended a 50% reduction of N and P fluxes delivered to the Greater North Sea with respect to the reference year 1985. For rigorously assessing the degree of achievement of this recommendation, the actual fluxes in 2007 cannot be simply compared to those of 1985, because of the difference in hydrological regime for the two years. In our modelling approach, hydrological forcings can be separated from anthropogenic factors (land use and agricultural practices and wastewater management). We therefore calculated the nutrient fluxes at the outlet of the 3S using the point and diffuse nutrient sources of 2007 combined with the hydrology of 1985 (Table 4). The differences between the runs in 1985 and in 2007 using the hydrology of 1985 can be explained only by changes in anthropogenic factors. The increase or levelling off of N–NO_3^- delivered to the coast can be explained by the inefficiency of the measures taken against the agricultural diffuse pollutions. The decrease of N–NH_4^+ and of total P is due to the improvement in terms of urban and industrial waste waters treatment. In the case of the Scheldt basin, due to its dense urbanisation, the decrease of N–NH_4^+ explains the decrease observed in terms of N fluxes delivered to the coast. The results (Table 4) show that whereas the OSPAR objective was achieved for P, the N flux in 2007 was higher (or still high) than in 1985 (1875 kgN km^{-2} vs. 1530 kgN km^{-2} for the Seine and 1845 kgN km^{-2} vs. 1550 kgN km^{-2} for the Somme, 1390 kgN km^{-2} vs. 1905 kgN km^{-2} for the Scheldt) at the same hydrology.

5.5. Effects on the coastal zone

The response of *Phaeocystis* blooms in the Belgian coastal zone (BCZ) to changing river nutrient inputs since 1984 were evaluated

Table 4
Calculated fluxes of nutrients at the outlet of the Seine, Somme and Scheldt rivers in 1985, 2007 and using the 2007 point and diffuse sources with the hydrology of 1985.

		N flux ($kgN\ km^{-2}\ yr^{-1}$)	P flux ($kgP\ km^{-2}\ yr^{-1}$)	Si flux ($kgSi\ km^{-2}\ yr^{-1}$)	N/P	N/Si	P/Si
1985	Seine	1530	290	865	5.28	1.77	0.34
	Somme	1550	85	1600	18.24	0.97	0.05
	Scheldt	1905	210	1050	9.07	1.81	0.20
2007	Seine	1830	90	820	20.33	2.23	0.11
	Somme	1845	55	1320	33.55	1.40	0.04
	Scheldt	1390	110	1100	12.64	1.26	0.10
2007 with hydrology 1985	Seine	1875	93	710	20.16	2.64	0.13
	Somme	2200	70	1580	31.43	1.39	0.04
	Scheldt	1100	100	970	11.00	1.34	0.10
Evolution from 1985 to 2007 (%)	Seine	+19.7	−68.9	−5.3			
	Somme	+19.0	−33.3	−17.5			
	Scheldt	−27.1	−48.7	+4.6			
Evolution from 1985 to 2007 using the hydrology of 1985 (%)	Seine	+22.7	−67.7	−17.8			
	Somme	+41.7	−20.3	−1.3			
	Scheldt	−42.3	−51.5	−7.8			

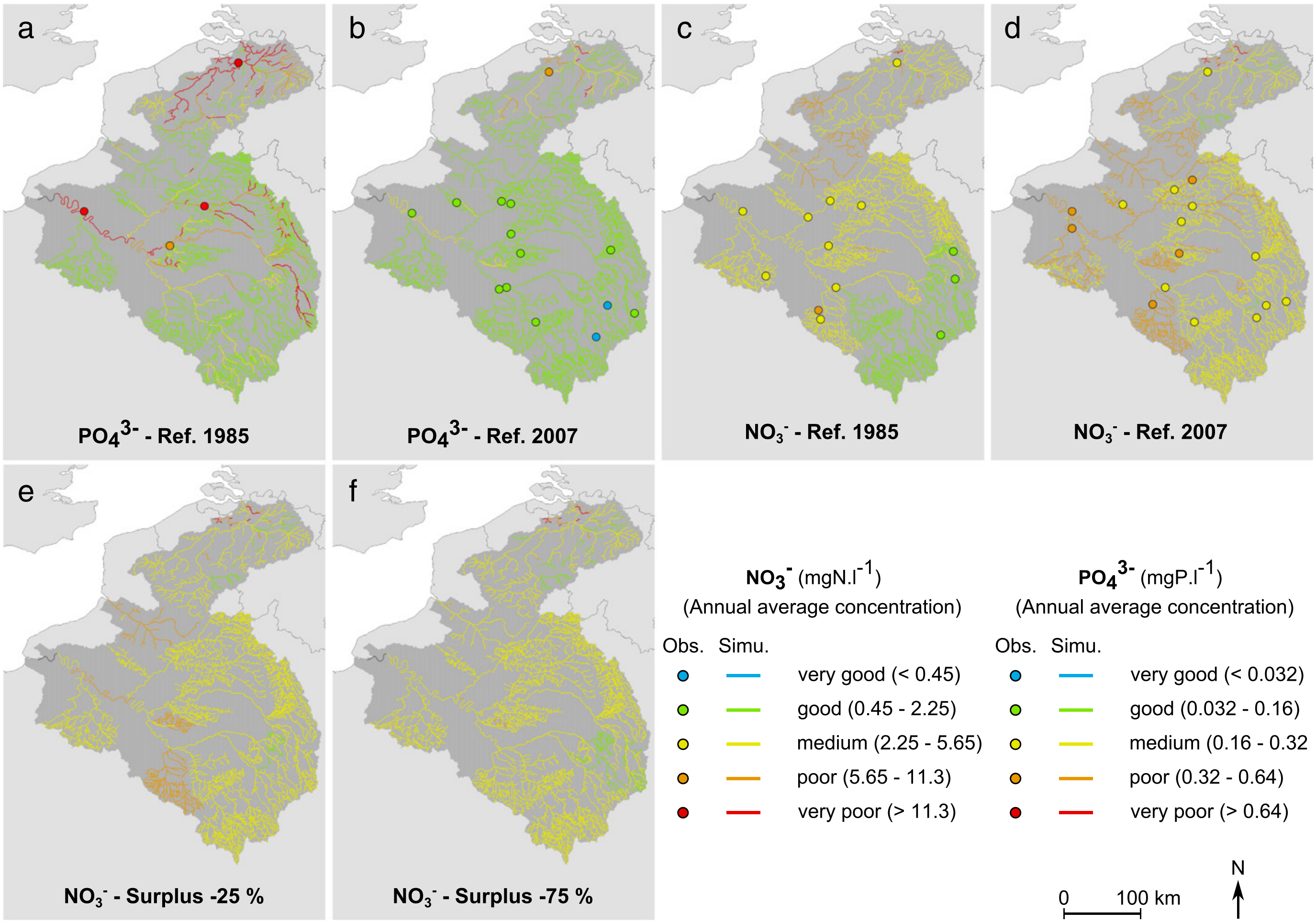

Fig. 10. P–PO_4^{3-} annual average concentrations within hydrological networks in 1985 (a) and 2007 (b), N–NO_3^- annual average concentrations within hydrological networks in 1985 (c) and 2007 (d) and with 25% (e) and by 50% (f) surplus decreases.

with the MIRO multi-box model forced with prevailing meteorological conditions and SR simulations of nutrient fluxes delivered by the Seine, Somme and Scheldt rivers. The results in terms of nutrients in the Belgian Coastal Zone are shown on Fig. 12. Although the simulation is not perfect, it catches the general trend of the major nutrients. The maximums of nitrate tend to decrease from the early 2000s (Fig. 12 a), while phosphate decreased from the middle of the nineties (Fig. 12 b). The silica does not show any trend (Fig. 12 c) and the maximums of the total chlorophyll a slightly decreased since the early 2000s (Fig. 12 d). The results (Fig. 13) are also appraised as maximum *Phaeocystis* colony cells reached (10^6 cells l^{-1}) and bloom duration (days), making use of the ecological indicator of 4 10^6 cells l^{-1} determined by Lancelot et al. (2009) for scaling undesirable *Phaeocystis*. The simulated long-term evolution of *Phaeocystis* maximum abundance (Fig. 13 a) compares reasonably well with observations, although calculated peaks are often higher than observations, due to the weak temporal resolution of observed data. For instance, the observed high values in 1993 only last 8 days, while in average only 9 to 11 observed data are available per year. Nevertheless, simulation shows some 50% reduction over the period, i.e. from 60 to 30 $\cdot$ 10^6 cells l^{-1}. Using the conversion factors described in Lancelot et al. (2005), this would correspond to a chlorophyll decrease over the simulated period from 34 to 17 mg m^{-3}. Despite this significant decrease, the maximum *Phaeocystis* abundance still remains well above the threshold value of 4 $\cdot$ 10^6 cells l^{-1} corresponding to a healthy ecosystem characterised by an efficient transfer of *Phaeocystis* production to higher trophic levels as defined by Lancelot et al. (2009). Over the period, the duration of undesirable *Phaeocystis* blooming defined as the number of successive days with abundance greater than 4 $\cdot$ 10^6 cells l^{-1} (Lancelot et al., 2011) decreased from 51 to 25 days (Fig. 13 b). Overall nutrient reduction measures taken on the 3S watersheds over the last two decades reduced undesirable *Phaeocystis* blooms by 50%.

The link between nutrient river enrichment and *Phaeocystis* blooms is evidenced in Fig. 13 c. On this figure the SR–MIRO annual production of *Phaeocystis* colonies in the BCZ is plotted against the calculated total annual P fluxes to the zone, directly from the Scheldt, and from Atlantic inflows enriched by the nutrient inputs of the Seine and the Somme rivers, in excess over the corresponding silica flux, taking into account the Redfield ratios (as already discussed by Lancelot et al., 2011). This excess P is expressed in terms of the potential biomass of non siliceous algae which can be produced on it. This analysis is based on the assumption that diatoms are growing first, up to the exhaustion of silica, the remaining nutrients being then used for the growth of non siliceous algae (Billen et al., 2007b; Officer and Ryther, 1980). The diagonal represents the situation where all the P in excess over silica is transformed into *Phaeocystis* biomass, expressed in carbon unit using the Redfield ratio (Fig. 13 c). In the eighties, a large part of the P was not transformed into biomass, suggesting that N was the limiting nutrient. On the contrary in the late 2000s years, almost all the P was transformed into biomass, showing that P becomes the limiting nutrient. The results show that *Phaeocystis* annual production is controlled by P ($r^2 = 0.63$; Fig. 13 c) loads rather than N, which is always in excess with respect to P or Si. This is further supported when relating the annual *Phaeocystis* production to the

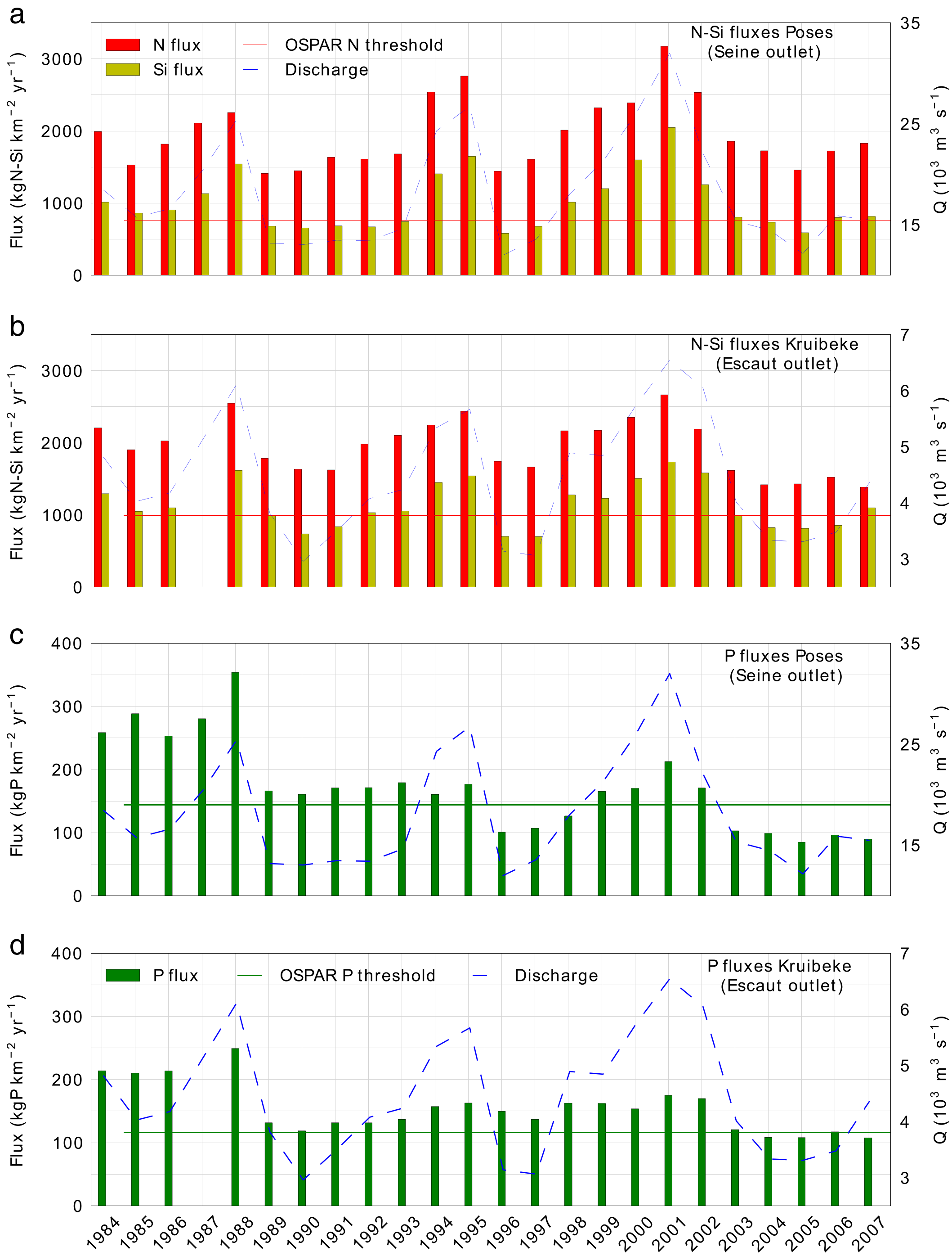

Fig. 11. SR-simulated N, Si and P annual specific fluxes delivered to the sea at Poses (a, c) and Kruibeke for the period 1984–2007 (b, d) (no hydrological constraints available for the Scheldt River in 1987).

potential eutrophication indicator ICEP (Garnier et al. 2010) estimated from SR–MIRO N, P, Si loads to the BCZ. This indicator is primarily based on nutrient requirements by diatoms (molar C:N:Si:P = 106:16:16:1; (Brzezinski and Nelson, 1995; Redfield et al., 1963), the excess N or P being converted to carbon as an estimate of primary production associated with agricultural eutrophication. Due to the P control of *Phaeocystis* production, the ICEP indicator is better expressed based on P excess. As shown in Fig. 13, the long-term trajectory of *Phaeocystis* production closely follows the reduction of P inputs. Moreover, the *Phaeocystis* production quantitatively matches the P excess brought into the zone, leaving only 20–30% unutilised. No such link between *Phaeocystis* production and N input is observed. N, greatly in excess over P, is not a limiting nutrient for *Phaeocystis* production over the whole period of study. This means that a large part of N input into the BCZ is exported to the North-East, where it can have further eutrophication effects.

6. Discussion

Like many river systems in densely populated and intensively cultivated regions of the world, the Seine, Somme and Scheldt rivers have been considerably enriched in both P and N with respect to the still nearly natural Si level (Howarth et al., 1996). When the OSPAR convention defined its aim of reducing by 50% (compared to

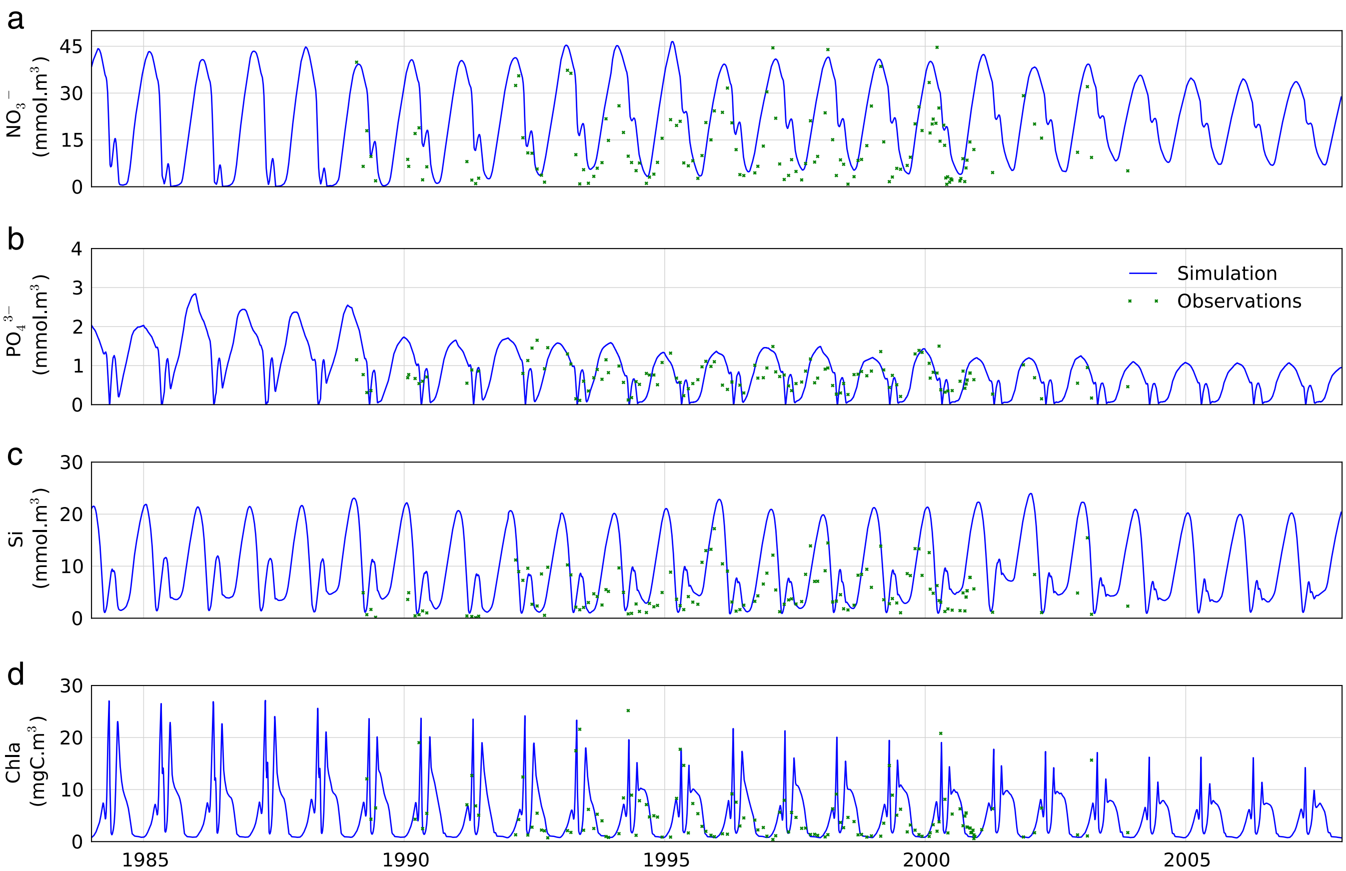

Fig. 12. Simulation of NO^{3-} (a), PO_4^{3-} (b), silica (c) and total chlorophyll a (d) in the Belgian Coastal Zone.

1985 levels) the P and N delivery from land-based sources to the sea, both nutrients were in excess over Si, which resulted in severe eutrophication problems in the French and Belgian coastal waters such as toxic algal episodes in the Seine Bight (Cugier et al., 2005) and *Phaeocystis* foam accumulation in the Northern French, Belgian, Dutch and German coasts (Lancelot et al., 1987; Skaloud et al., 2006). Severe deterioration of freshwater quality within the drainage network of the 3S rivers was also caused by excess P and N inputs from the watershed, including eutrophication of large river and stagnant areas, oxygen depletion and generalised NO_3^- contamination. The measures taken by water authorities in compliance with the urban wastewater European directives (Council Directive 91/271/EEC) first consisted in increasing the sewer connection rate and improving wastewater treatment. It helped to decrease the NH_4^+ discharge to rivers by 70% since the early 2000s, thanks to the implementation of a nitrification step (Even et al., 2007). The banishment of poly-phosphates from washing powders and the implementation of specific P removal steps in wastewater treatment led to a considerable reduction of urban P sources. The more recent and still ongoing implementation of denitrification in the largest WWTPs comprises the last phase of these efforts to reduce point sources of nutrients to surface water. Nowadays, point sources of P in the rivers studied here have been reduced to the same level as diffuse sources of this element from erosion of agricultural soils, and roughly 80% of N originates from diffuse sources (Thieu et al., 2009). The limit of the policies devoted to point sources of nutrients is therefore close to being reached in terms of environmental improvement.

These measures have considerably improved freshwater water quality as far as NH_4^+ and P contamination is concerned. The OSPAR objective of reducing P delivery to the coastal zone by 50% has been achieved. As a result, while N controlled *Phaeocystis* colony blooms in the 1980s, it is now P that controls eutrophication in the BCZ, and both the intensity and the duration of *Phaeocystis* blooms have been reduced by half. This shift from N to P limitation was also shown in the Adriatic Sea impacted by the nutrient loads from the Po River (Cozzi and Giani, 2011).

On the other hand, as far as N is concerned, the situation has not improved in the past 24 years. NO_3^- contamination of ground and surface freshwater resources is still a matter of great concern. River N flux delivered to the coastal zone has not been significantly reduced in spite of the OSPAR recommendations. A large amount of excess N, not used by algal growth in the BCZ, is exported to adjacent areas to the North. This situation results from the fact that no or insufficient interventions have been devoted to reducing agricultural sources of nutrients, which clearly dominate over urban sources in terms of N inputs. We suggest that the agricultural soil N surplus might be a convenient indicator to assess the efficiency of measures aiming at limiting agricultural NO_3^- losses. Our analysis of N soil surpluses in the different agricultural areas of the 3S watershed shows that these have increased considerably since the middle of the 20th century, and that recent limited reducing trends are only observed in the Walloon and Flemish areas of the Scheldt basin, while the trend is still increasing in most areas of the Seine and Somme basins (Fig. 6). Efforts to reduce N contamination of hydrosystems should aim to reduce N leaching into rivers as managing landscapes (Passy et al., 2012) or reduce agricultural N soil surpluses (Lassaletta et al., 2012), either by precision agriculture techniques (Di and Cameron, 2002; Tilman et al., 2002), good agricultural practices (Thieu et al., 2010) or by organic farming (Billen et al., 2012; Gypens et al., 2013 this issue; Thieu et al., 2011). To illustrate the potential sensitivity of the system to such measures, we ran the SR–MIRO chain model for a scenario representing the 2007 conditions with a 25% and a 50% reduction of

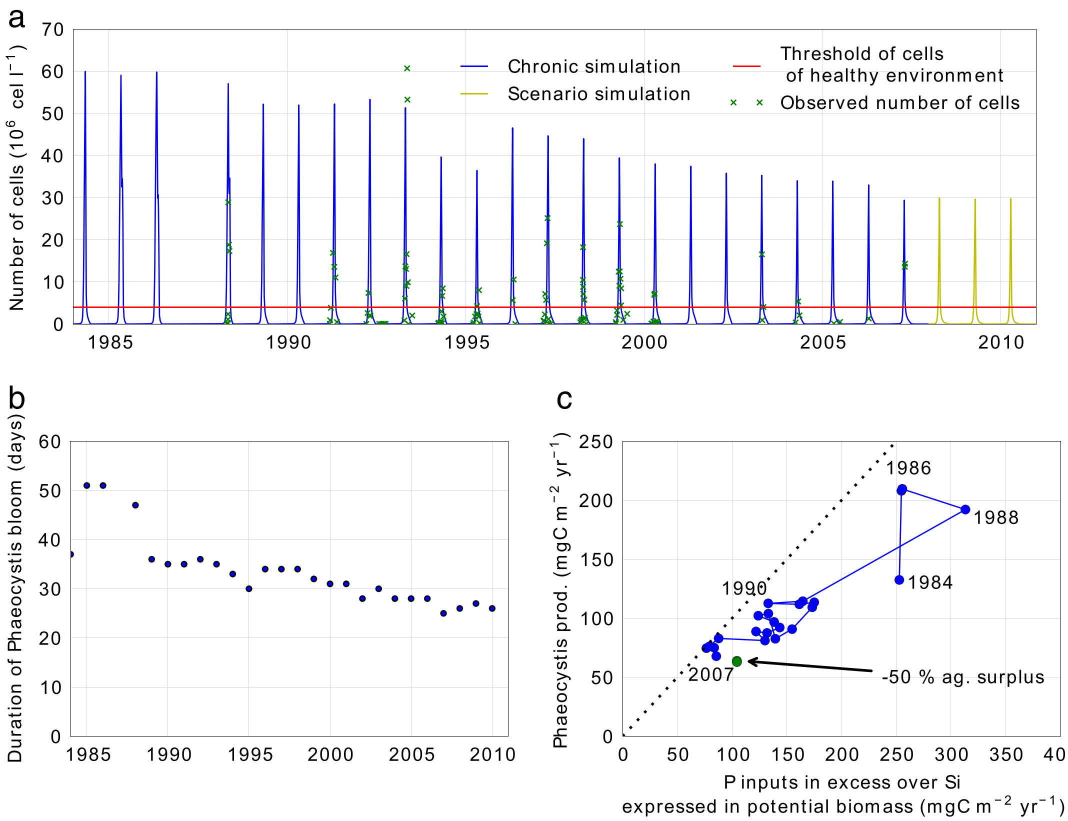

Fig. 13. Abundance of *Phaeocystis* over the 1984–2007 period and for the two scenarios explored in the BCZ in 10^6 cells l^{-1} (a); duration of the *Phaeocystis* blooms for the period studied and for the two scenarios explored, in days (b) and production of *Phaeocystis* expressed in potential biomass in mgC m^{-3} yr^{-1} as a function of excess P input over silica (c). The red line represents the threshold of cells of *Phaeocystis* corresponding to a healthy marine ecosystem (Lancelot et al., 2009).

the agricultural soil surplus of all agricultural land, and assuming an immediate response of aquifers. The results show a significant decrease of NO_3^- contamination of rivers, with most waterbodies of the three watersheds reaching NO_3^- concentrations less than 5.65 mgN–NO_3^- l^{-1}, half the standard for drinking water quality (Fig. 10 e, f). In the scenario with 50% reduction of the agricultural surplus, the flux of N delivered to the sea would be reduced to 1215, 1070 and 1200 kgN km^{-2} yr^{-1} for the Seine, Somme and Scheldt, respectively, which would meet the OSPAR target. The simulation shows, however, that no further significant effect on *Phaeocystis* blooms would be achieved, in terms of neither intensity nor duration, because N remains in excess over P, which is still controlling the algal dynamics (Fig. 13). Further limitation of *Phaeocystis* blooms will thus require additional actions to reduce P inputs to the sea, including measures addressing the diffuse sources of this element.

7. Conclusion

Nutrients transfer through the watersheds of the Seine, Somme and Scheldt greatly changed from 1984 to 2007. In 1985, the OSPAR reference year, the excess of P was the major problem. Within the hydrological networks, only the upstream parts had P concentrations less than 0.032 mgP l^{-1}. Fluxes delivered to the coastal zone reached 290 kgP km^{-2} yr^{-1}, 85 kgP km^{-2} yr^{-1}, and 210 kgP km^{-2} yr^{-1} for the Seine, Somme and the Scheldt respectively. The decrease of P fluxes was more than 50% from 1985 to 2007, so the OSPAR P objective is reached. Concerning N–NO_3^-, the upstream parts of the hydrological networks had N concentrations less than 2.25 mgN–NO_3^- l^{-1}. But two thirds of the drainage network had N concentrations ranging from 2.25 to 5.65 mgN–NO_3^- l^{-1}. In terms of fluxes at the coastal zone, 1530 kgN km^{-2} yr^{-1},1550 kgN km^{-2} yr^{-1} and 1905 kgN km^{-2} yr^{-1} were delivered by the Seine, Somme and the Scheldt respectively. N was the limiting nutrient for *Phaeocystis* development, and blooms achieved more the 60 · 10^6 cells per litre and lasted more than 50 days.

From 1985 to 2007, thanks to the improvement of the waste water treatment, point sources pollution greatly decreased. P, mostly originating from point sources, drastically decreased during this period. Almost all the rivers of the three watersheds have now P concentrations below 0.16 mgP l^{-1}. Only the Seine from Paris to the estuary and sectors from intermediate rivers to main lower branch of the Scheldt Rivers have P concentrations above 0.16 mgP l^{-1}. P fluxes delivered to the coastal zone also greatly decreased. In 2007, they are 90 kgP km^{-2} yr^{-1} 55 kgP km^{-2} yr^{-1} and 110 kgP km^{-2} yr^{-1} for the Seine, Somme and Scheldt respectively. Per contra, N–$NO_3{}^-$ concentrations in rivers increased from 1985 to 2007. At this date, the N–$NO_3{}^-$ concentrations are ranging from 0.16 to 0.32 mgN–$NO_3{}^-$ l^{-1}, in almost the two thirds of the rivers of the three watersheds. In the Loing, Somme and Eure basins, these concentrations are above 0.32 mgN–$NO_3{}^-$ l^{-1}. It is also the case for some parts of the Scheldt River and the downstream part of the Seine River. In 2007, 1830 kgN km^{-2} yr^{-1}, 1845 kgN km^{-2} yr^{-1} and 1390 kgN km^{-2} yr^{-1} are delivered to the coastal zone by the Seine, Somme and Scheldt respectively, the OSPAR N objective being far from reached. The same trends of P and N fluxes were reported in many places in Europe (Bouraoui and Grizzetti, 2011; Grizzetti et al., 2012).

As a whole, because of its sharp decrease, P is now the limiting nutrient in the coastal marine waters, driving the *Phaeocystis* blooms decrease, in amplitude and duration. On the other hand, the suspected role of excess N in the induction of algal toxicity in marine systems, particularly in the case of domoic acid production, pleads for a reduction of N fluxes delivered to the sea (Klein et al., 2010; Trainer et al., 2012).

This paper has illustrated the potentialities of a modelling approach coupling a watershed model with a coastal sea model to analyse the long-term trends of freshwater quality, river fluxes delivered to the sea and the response of the marine ecosystem. This type of approach is required to better target the future actions aiming at controlling water quality in the aquatic continuum from freshwater to coastal zones.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jmarsys.2013.05.005.

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