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Modeling historical changes in nutrient delivery and water quality of the Zenne River (1790s–2010): The role of land use, waterscape and urban wastewater management

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A B S T R A C T

The Seneque/Riverstrahler model has been used to explore the effect of human-induced changes in drainage network morphology and land use on organic and nutrient pollutions, for the last 20 years and back to the 1890s and 1790s. With the development of human civilization, past environmental constraints differed compared to today. Research has sought to reconstruct (i) point sources (domestic and industrial), using statistics and archives from these periods, and (ii) diffuse sources via landscape and riverscape analysis based both on maps and agricultural statistics from the periods concerned.

This study shows that a maximum of pollution occurred in the 1890s at the height of the industrial period, due more to the industrial load than to the domestic load. This substantial organic and nutrient pollution might have lasted up to very recently, when the Brussels Northern wastewater treatment plant began operation in 2007, significantly reducing the organic and nutrient load of the Zenne River, returning to a background pollution level assessed herein for the 1790s before industrialization expanded.

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1. Introduction

An increasing population with its urbanization and sanitation needs, as well as changes in industrialization and agricultural practices and structural landscape transformations have considerably modified the natural biogeochemical cycles of essential bio-elements from local to larger scales (Billen and Garnier, 1997; Sutton et al., 2011).

As a result, nitrogen, phosphorus, and carbon inputs from land-based sources to surface waters have largely increased from the early industrial period to the late 1990s and early 2000s, leading to organic pollutions and subsequent anoxic conditions as well as eutrophication problems in surface fresh- and marine waters. Recently however, the application of several environmental policy directives, for example the European Water Framework Directive (EU-WFD 2000/60/CE), mainly targeting urban point sources, led to massive decreases of carbon, ammonium and phosphorus emissions to the watersheds and better oxygenation of surface waters (Bouraoui and Grizzetti, 2011; Garnier et al., 2007). As far as diffuse agricultural sources are concerned, however, the measures taken so far, although they often prevented a further increase of nitrogen contamination, failed to produce a significant decrease (Billen et al., 2005; Bouraoui and Grizzetti, 2011). Such changes at any location in the watersheds have an influence not only on the surrounding biogeochemical functioning of surface freshwaters (namely river systems and their stagnant annexes), but also on the parts of the aquatic continuum farther downstream: large rivers, estuaries and coastal seas (Boyer et al., 2002; Cloern, 2001; Lancelot et al., 2011; Rabalais et al., 2009).

The Zenne River, a tributary of the Scheldt River via the Dijle and Rupel Rivers, represents an emblematic case study for understanding and modeling the importance of organic and nutrient loads coming from highly populated watersheds, and the transfer of pollution through the drainage network to the estuarine and coastal marine zones. Despite its small size and modest water contribution (7%) to the Scheldt and Belgian coastal zone, it has been calculated (Passy et al., 2013-this issue; Thieu et al., 2009) that the Zenne contributes today up to 25% of the total N and P delivered by the Scheldt to the North Sea.

An interesting feature of the Zenne is that since the Middle Ages, the river played a major role in the development of the city of Brussels and consequently its hydromorphology and pollution are inherently linked to the city's development (Billen and Duvosquel, 2000; Deligne, 2003). It is interesting to note that historically, due to the asymmetry of the

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river bank morphology in the Brussels area, human activity essentially developed on the flat left bank, whereas the hilly right bank developed into a residential area for the aristocracy (Billen and Duvosquel, 2000). Early in history, the Zenne River's hydrology was considerably modified (mill operation, navigation, etc.), to such an extent that Brussels was called "Portus" at the beginning of the 11th century (Deligne, 2003). Similar to other famous medieval cities of Northern Europe, a prosperous textile craft industry strongly contributed to the demographic, economic and cultural expansion of Brussels, soon supported by the construction of the Willebroeck canal linking Brussels to Antwerp, at the estuary, in the 16th century (Deligne, 2003). In the 19th century, due to (i) the natural increase in alluvium and (ii) water withdrawn by the new Charleroi canal, the Zenne discharge decreased, which further enhanced alluvium and reduced the dilution of pollution coming from craft industries; as a result, for sanitation purposes, the river was covered in the 19th century (a work which lasted from 1867 to 1871, Demey, 1990), while most mill ponds had already disappeared a century before (Deligne, 2003; 2012). Since the river disappeared from the perception of riverine inhabitants, the preservation of water quality was no longer a priority for Brussels, even though, in the late 20th century, most other larger cities were equipped with efficient wastewater treatment plants (WWTPs). The implementation of national directives, but most particularly the European directives (EU-Urban waste water treatment directive, UWWTD, 1991; EU-Water framework directive, WFD, 2000) pushed for the improvement of wastewater treatment plants (WWTPs) or construction of new ones. However, despite its status as a European city and the European Union capital, Brussels was paradoxically only very recently equipped with WWTPs (in 2000 and 2007 for the Brussels South and North WWTPs, respectively).

The present study aims at highlighting, over a long historical period of two centuries (1790s, 1890s and the recent period of 1990–2010), the particular role played by Brussels on the Zenne River's ability to transfer pollutants to the Scheldt estuary and North Sea coastal zone. Aware of huge industrial activities in the past, with a maximum in the 1890s (Billen et al., 1999), we wished to explore the importance of these industrial sources in terms of organic matter and nutrient contamination of surface waters.

The working strategy consisted first in quantifying the changes in water quality in the modern period, 1990–2010, from before to after WWTP construction for the Brussels conurbation, using a GIS-based biogeochemical modeling approach (Seneque/Riverstrahler, Ruelland et al., 2007). After validation on this recent, well-documented period of significant changes in urban organic loading, the model was adapted and applied to much earlier historical periods in order to place the recent changes within a broader historical perspective. The results of a previous interdisciplinary work (Billen et al., 1999), evaluating the early (circa 1890) industrial and domestic loads, were used for this purpose, whereas the ones for 1790s were extrapolated based on demography data.

2. Site description

The Zenne watershed has a surface area of 1160 km^2 and is a small order 3 tributary of the Rupel, a major basin of the Scheldt (Table 1). Due to the proximity of the marine coastal zone, the climate is rainy, with annual precipitation amounting to 850 mm, and mild, with a mean temperature of 15 °C; the topography is flat with an altitude of 140 m at the spring and a mean slope of 0.23%.

The main branch of the river extends over 58 km between Lembeek and Zennegat at the confluence with the Dijle River and its downstream part belongs to the tidal Scheldt estuary.

For the last 20 years, the Zenne River at Lembeek has had an average discharge of 3.8 $m^3 s^{-1}$ and, most important, receives a similar amount (3.5 $m^3 s^{-1}$) of urban effluents when crossing the city of Brussels; the contribution of Brussels urban waters is enormous compared to the river water discharge. During summer low discharges, urban effluents can be even higher than the river discharge. During rain events, besides treated water from the wastewaters, urban surface runoff water and untreated water may overflow in the Brussels area (see below). The Zenne River at Eppegem has had an average discharge of 10 $m^3 s^{-1}$ for more than the 20 past years, rather low compared to 62 $m^3 s^{-1}$ and 140 $m^3 s^{-1}$ for the Rupel and Scheldt, respectively.

The Zenne River crosses the Brussels conurbation over a distance of about 20 km starting 19 km after Lembeek. The proportion of urban area of the Zenne is the highest in the Scheldt basin and presently reaches 40% of the land use, with forest occupying the lowest proportion. The population density exceeds 1200 inh km^{-2}, more than twice higher than that of the whole Scheldt basin (Table 1).

The Zenne basin is characterized by 13 major sub-basins, including the upstream Zenne watershed (Zenne WSH, Fig. 1, Table 2) and a main axis from Lembeek to the confluence with the Dijle River, Eppegem being the last measurement station, 41.5 km from Lembeek, where the tidal zone starts (Fig. 1).

As analyzed later in the paper, in order to run the model for historical situation, we documented the constraints that have necessarily changed in the Zenne basin. With a population that increased by a factor of 35 during the past two centuries, and with a flourishing industry early in the history, point sources were logically expected to be strongly modified. To quantify them, we made use of archives. For land use changes, old maps were digitalized and compared to the recent Corine land cover data bases (1990, 2006). Historical agricultural nitrogen budget was also established so that diffuse sources could be evaluated along the studied period. Regarding the geomorphology of the drainage network, we made the assumption that the presence and connectivity of ponds were the major difference in the hydrological landscape between the end of 1700s and today.

3. The modeling approach

The biogeochemical modeling approach, in development since the 1990s (Billen et al., 1994; Garnier et al., 1995), has recently been coupled to a GIS interface (Ruelland et al., 2007), allowing a spatial description of drainage networks and their environmental constraints within their watersheds. The field of implementation of the modeling tool, initially dedicated to the Seine River (France), has been extended to several other drainage basins, differing in their size, their hydroclimatic region and their degree of human impacts. The Seneque/Riverstrahler model was recently implemented on the entire Scheldt drainage network (Passy et al., 2013-this issue; Thieu et al., 2009), focusing on contrasted hydrological years in the 2000s and on a reconstruction of the water quality for the past 30 years, with no particular attention paid to

Table 1
Major characteristics of the Zenne basin. Comparison with its increasing embedding basins for the present period (2000s). Inh. for inhabitants.

Sub-basins and basin	Watershed area, km^2	Average discharge, $m^3 s^{-1}$	% Crop land	% Grass and heterogeneous land	% Forest	% Urban	Population, Inh.	Population density, Inh. km^{-2}
Zenne	1200	10	30.3	22.9	6.5	40	1,546,931	1289
Rupel	6678	62	23.8	32.2	11.8	30.1	4,062,509	608
Scheldt	19,900	140	39.4	24.1	7.7	25.1	10,746,344	540

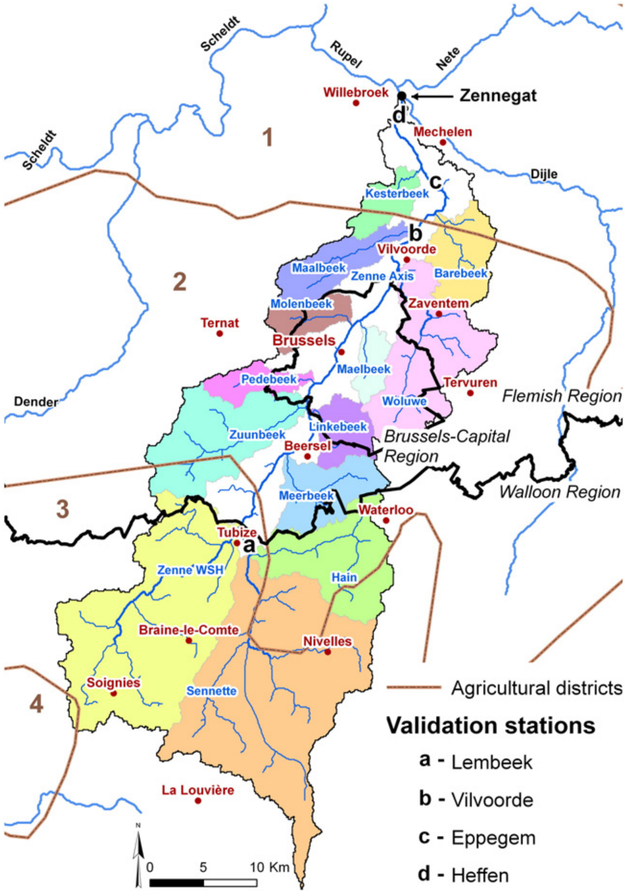

Fig. 1. The hydrographic network of the Zenne River and associated sub-basin units. Main cities and major stations with observation data used for validation along the Zenne axis are indicated. The various colors allow differentiating the 12 tributaries of the Zenne, taken into account as inputs to the main branch. The main branch of the Zenne River receives in addition the water from its own watershed (in white). The brown lines distinguish the major agricultural districts used to differentiate diffuse sources (from top to bottom: 1. Sandy; 2. Sandy–loamy; 3. Loamy; 4. Hainaut Campine). The thick black line separates the Flemish and Walloon regions; Brussels region is also pointed out by a black polygon.

the specificities of the Zenne River. The model has also been applied to fecal bacteria modeling (Ouattara et al., 2013-this issue).

The implementation of the model on any watershed requires the elaboration of constraint databases on the hydromorphology (GIS layers of river channels and their watersheds, stagnant pond systems and discharge data), point sources (domestic and industrial loads) and diffuse sources (land use). Once these data are gathered, water discharges are routed through the drainage network, divided into sub-basins and main axes, depending on the spatial resolution required, and coupled with an ecological model (the RIVE Model, see Garnier et al., 2002). The RIVE model includes the exchanges of nutrients (N, P, Si), organic matter and oxygen between the biological compartments of phytoplankton (three functional groups), zooplankton (two functional groups), bacteria (heterotrophic and nitrifying bacteria), suspended solids and associated phosphate reversible adsorption, and a benthic module of nutrient exchanges across the sediment–water interface as a result of a given sedimentation flux of organic material (Thouvenot et al., 2007). The resolution of the output state variables is 10 days, although calculations using biological kinetics are done at a sub-hourly time step.

The constraint databases for the Zenne system were implemented as follows.

3.1. Geomorphology

The drainage network was established from the SRTM DEM (Digital Elevation Model, NASA, 2000) at a resolution of 90 m, and divided into 13 sub-basins and one branch (from Lembeek to the confluence with the Dijle, with a resolution of 1 km, Fig. 1). The slope, length and width of the rivers were evaluated according to Thieu et al. (2009). The model calculates depth from the discharge, width and slope of each river stretch using standard hydraulic relationships, as also described by Thieu et al. (2009).

To take into account the fact that the Zenne was covered from the late 1860s (1867–1871), in the city of Brussels, model runs were performed for the last 140 years applying no solar irradiance to the covered river stretch (20 km in Brussels area). Regarding the hydrological annexes (see Table 4), a majority of the ponds are today sand-pit lake used for extracting building material and as recreational area, having lost their connectivity with the drainage network. Differently from those in the historical period when they were directly impounded within the river bed, ponds are not taken into account in the modeling simulations for the period 1990–2000.

Hydrology: To run the model, the daily discharges of the period from 1990 to 2010 were selected (Fig. 2). The water fluxes were partitioned into a direct runoff and base flow component, using the recursive digital filter method (Eckhardt, 2005; 2008) based on the observed daily values at Eppegem (data from Hydrologisch Informatie Centrum, Waterbouwkundig Laboratorium, Departement Mobiliteit en Openbare Werken van de Vlaamse Overheid; http://www.waterstanden.be/), the outlet station of the hydrological network.

Table 2
Change in land use in the Zenne basin for the years 1990 and 2006 (Corine land cover). Zenne WSH is the upstream sub-basin of the Zenne (see Fig. 1).

Sub basins and basin	Watershed area, km^2	% Crop land		% Grass and heterogeneous land		% Forest		% Urban		Total basin	
		CLCI990	CLC2006	CLCI990	CLC2006	CLCI990	CLC2006	CLCI990	CLC2006	CLCI990	CLC2006
Zenne branch	192	12	11	22	22	2	2	63	64	99	99
Zenne WSH	236	47	46	31	31	4	4	17	18	100	100
Sennette	272	46	45	25	25	7	8	22	23	100	100
Hain	85	38	37	15	13	9	9	38	40	100	100
Zuunbeek	91	32	32	43	43			25	25	100	100
Linkebeek	28	4	4	8	8	7	7	80	80	100	100
Pedebeek	22	14	14	45	45			41	42	100	100
Molenbeek	25	4	4	14	13	1	1	81	82	100	100
Maelbeek	17							100	100	100	100
Woluwe	103	12	12	2	2	19	18	67	69	100	100
Maalbeek	42	29	29	23	22			47	48	100	100
Kesterbeek	23	36	35	33	35	5	5	24	25	99	99
Barebeek	47	35	35	12	12	12	11	41	42	100	100
Meerbeek	48	11	11	19	19	20	20	50	50	100	100

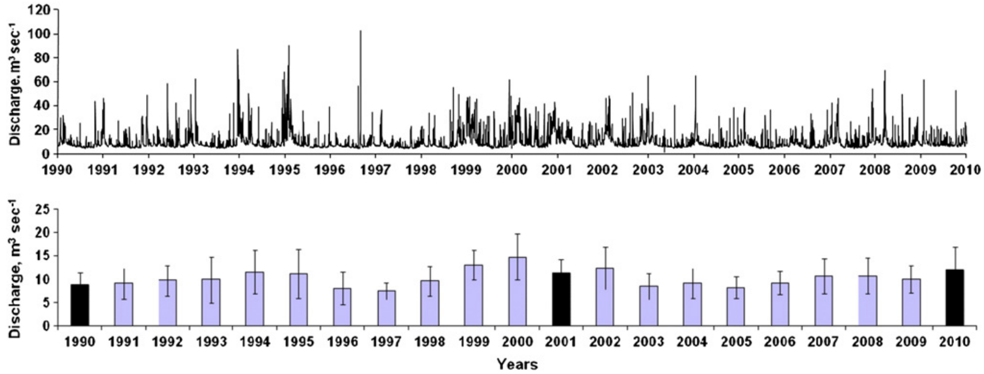

Fig. 2. Seasonal variations of the daily discharges (top) of the Zenne River (Eppegem), from 1990 to 2010. Mean annual discharges (bottom) at the same station (Eppegem) with standard deviations. Black bars show the years chosen in the data series for modeling purpose.
Data sources: Hydrologisch Informatie Centrum, Waterbouwkundig Laboratorium, Departement Mobiliteit en Openbare Werken van de Vlaamse Overheid; http://www.waterstanden.be/.

Distinguishing these two components of the discharge specifically takes the inputs of diffuse sources into account, through runoff and base flow, as well as the surface point sources and their transformation and transfer in the drainage network.

3.2. Diffuse source and land use

To calculate diffuse sources of nutrients to the hydrographic network, the Seneque/Riverstrahler model assigns a yearly constant mean composition to surface runoff and base flow, according to land use in the watershed (see Billen and Garnier, 1999). Corine land cover, a database of land use coverage of the European territory, is available (see European Environment Agency, http://www.eea.europa.eu/) for the years 1990, 2000 and 2006, which we assumed were representative of the years selected for study: 1990, 2001 and 2010, respectively. The major classes of land use considered were forest, grassland and arable land (Table 2).

For each type of land use, a nutrient concentration must be assigned for each of the model's variables (e.g., N, P, Si, C, suspended solids (SS) of primary interest for the modeling approach) to the two components of the discharge, surface and base flow (see Passy et al., 2013-this issue; Thieu et al., 2009).

In order to characterize the major changes in agricultural practices for each of the three agricultural regions of the Zenne basin (see Fig. 1), a complete nitrogen budget of arable land was calculated from the agricultural statistics available, for the years chosen to study the present period (Belgium National Institute of Statistics (INS) data for 1990, 2000, 2006, see Passy et al., 2013-this issue). The budget compares total N output by exported crops and total input by fertilization (synthetic fertilizer and manure application), legume atmospheric N fixation and atmospheric deposition. Total N output by exported crops was calculated from production figures and converted into N using standard N content data. The annual N surplus is assumed to be diluted by the mean infiltrating water flux, thus allowing the sub-root nitrate concentration to be calculated. This concentration is assigned to surface runoff, while the nitrate concentration assigned to base flow is determined from available survey data on groundwater quality (Passy et al., 2013-this issue). Note that the information gathered for 2006 is considered valid for 2010.

3.3. Point sources

For the present period, wastewater treatment plants have been inventoried and geo-referenced at the scale of the Zenne basin, and their loading to the rivers calculated from per capita emissions data based on the type of treatment in the WWTPs and their effective capacities.

For the 2000s, these data were obtained from Water or Environmental Agencies of the Walloon and Flemish regions. For the 1990s, a reconstruction of the point sources was carried out on the basis of changes in population and its rate of connection to sewer systems within the Zenne watershed, the year of the implementation of each WWTP and some assumptions about the evolution of waste water treatment (see also Passy et al., 2013-this issue).

The pollution in terms of inhabitant-equivalents, suspended solids, total organic carbon, total phosphorus and nitrogen as taken into account in the model is given in Table 3. The figures include a mixture of untreated wastewater and urban surface runoff water that may overflow during rain events in the Brussels area, due to the insufficient capacity of some stretches of the old sewer system. Given that today industry wastes are either treated by their own or municipal WWTPs, the industrial load is not explicitly taken into account for the recent years.

4. Modeling the water quality during the past two decades — model validation

We selected the years 1990, 2001, and 2010 for which we were able to collect sufficient data to implement the model, both in terms of constraints (discharges, point and diffuse pollution) and validation data (data sources: Vlaamse Milieumaatschappij, Geoloket Waterkwaliteit – http://www.vmm.be/geoview/ – and GESZ research project, Impulse Environment initiative from the Brussels Institute for Research and Innovation), the major quality variables of the ecosystem considered here being oxygen, ammonium and nitrate, phosphate and total phosphorus. These three years are well adapted to studying the Zenne River status after the implementation of the Brussels South and North WWTPs in 2000 and 2007, respectively; the year 1990 corresponds to the situation preceding the implementation of any wastewater treatment.

It is interesting to observe the improvement in water quality downstream from Brussels in 2010, showing that the domestic effluents of the Brussels conurbation are now much better treated, as demonstrated by the better water quality at Vilvoorde and Eppegem downstream from Brussels. An improvement in water quality is also

Table 3
Total pollution load in the Zenne basin for the periods considered (including industrial and domestic loads).

Date	Inh. Equ., 10^3	kg SS day^{-1}	kg Org. C day^{-1}	kg TP day^{-1}	kg TN day^{-1}
1790	320	19,218	15,060	38	298
1890	3196	192,175	150,603	3813	29,849
1990	1028	79,574	23,910	1222	12,557
2001	1820	79,709	24,209	1431	14,405
2010	2513	27,334	9483	1047	8105

observed between 2001 and 2010 in terms of oxygen, ammonium and phosphorus due to the implementation of new WWTPs and improvement of existing WWTPs in the upstream part of the watershed (Fig. 3).

No improvement, however, is apparent between 1990 and 2001, although part of the Brussels urban sewage (approximately 1/3) has been treated since 2000 by the Brussels South WWTP (Fig. 3). We therefore plotted the values for 1999 (immediately before any treatments of the domestic effluents of Brussels), as well as for 2000 (during commissioning). The Brussels South WWTP clearly did not significantly improve the water quality of the downstream river water (see Fig. 5 at Vilvoorde and Eppegem). This can mainly be attributed to the fact that Brussels South only treats part of the Brussels urban wastewaters, which does not suffice to overcome the negative effects of the remaining release of untreated sewage, especially regarding the small size of the receiving river. Finally, it is only after 2007 with the implementation of the Brussels North WWTP that oxygen conditions have been restored and biological oxygen demand (BOD) decreased; ammonium and total phosphorus values observed in the downstream river section decreased too. The behavior of nitrate shows, up to the 2000s, a substantial decrease by denitrification in the river downstream from the impact of the Brussels pollution load compared to the upstream station, due to an oxygen deficit, whereas after 2007 with oxygenation improvement, the difference between the upstream and downstream stations is smaller, e.g. in 2010 (Fig. 3).

We validated the model by comparing simulated longitudinal profiles with observations for periods when water quality observations exist (profiles, covering all seasonal situations, for the years 1990, 2001 and 2010, respectively) (Fig. 4). The agreement between the observations of quality variables and the model's calculations is quite good, especially for 2010, as validated with more variables (see Fig. 4C, D); also, the constraints data are better documented for the more recent period, especially those of point sources (Ouattara et al., 2013-this issue). Nevertheless, for the years 1990 and 2001, the major trends of the observations are well represented by the calculations (Fig. 4A, B).

Simulations of oxygen concentrations along the main branch of the Zenne River clearly represent the observed deficit in the Brussels area in 1990, before the implementation of the WWTPs, as well as an improvement after, in 2010 (Fig. 4A and C). In parallel, the spatial trends of the increase in ammonium, total phosphorus and phosphate in the urbanized area are well modeled; the levels of these variables are much higher for the years 1990 and 2001, compared to 2010 (Fig. 4). Interestingly, denitrification related to anoxia, substantially lowering the nitrate concentration, is also well represented by the model (Fig. 4). In addition to the good agreement of the model results with the observations obtained for 2010 for the variables routinely measured to assess organic pollution, others such as dissolved and particulate organic carbon (DOC and POC), phytoplankton biomass (Phy, expressed as chlorophyll *a* concentrations), silica (Si), required by the diatoms as a nutrient complementary to phosphorus and nitrogen,

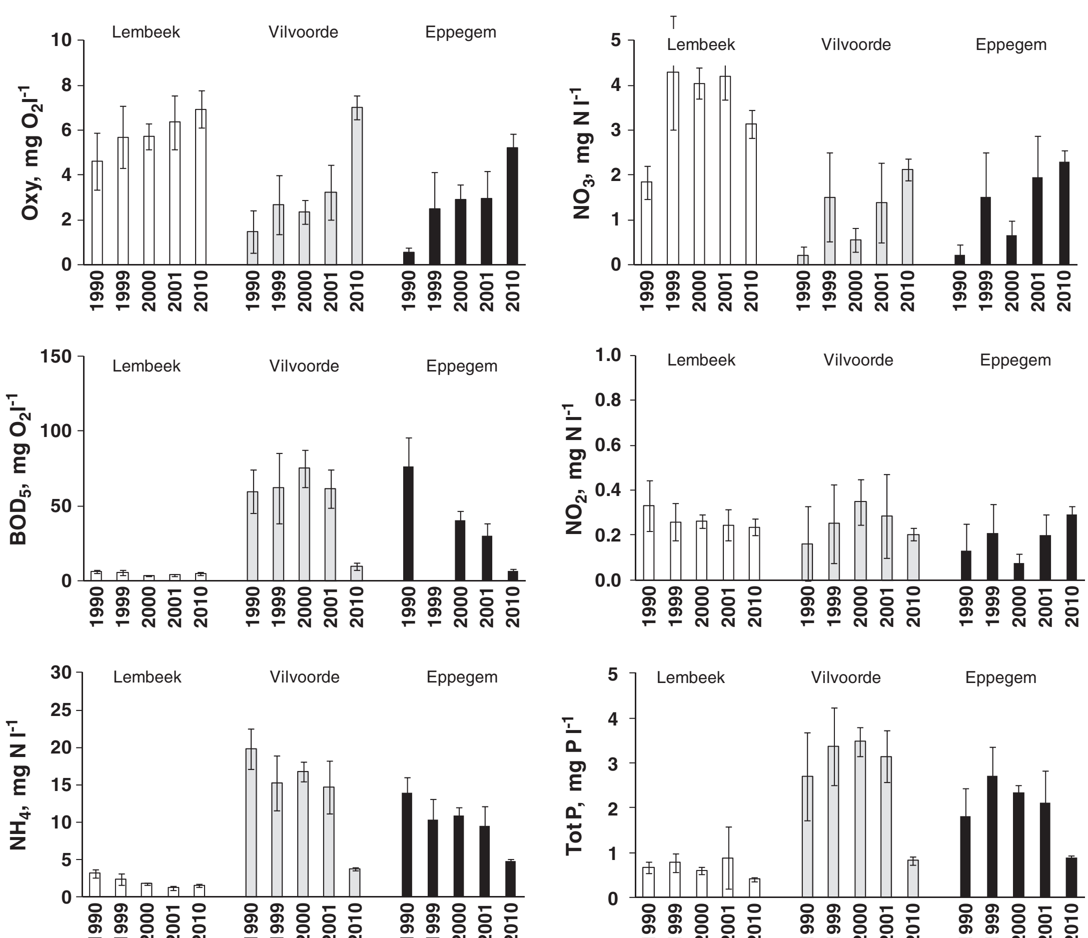

Fig. 3. Mean changes in major variables of water quality among the chosen years (1990, 1999–2000–2001 and 2010). These changes are shown for one station upstream of Brussels conurbation (Lembeek) and two downstream stations (Vilvoorde and Eppegem) (see Fig. 1). 2000 is the year of implementation of the first wastewater treatment plant for Brussels conurbation. Data sources: Vlaamse Milieumaatschappij, Geoloket Waterkwaliteit – http://www.vmm.be/geoview/ – and GESZ research project, Impulse Environment initiative from the Brussels Institute for Research and Innovation.

and suspended solids (SS) are also clearly in accordance with the observations (Fig. 4D).

This level of agreement between observations and simulations is far from trivial. The kinetics parameters were determined independently, either experimentally or based on the literature, and have already been validated for a number of watersheds. In addition to the temperate watersheds of the Seine, Somme and Scheldt (see Thieu et al., 2009 and references therein), the Red River in Vietnam in a monsoon subtropical area (Le et al., 2010) and the cold Swedish Kalix and Lule rivers (Sferratore et al., 2008) were validated. Therefore, this agreement means that the constraints have been correctly established.

5. Exploring the water quality of the Zenne in earlier historical periods

To model the Zenne system under historical conditions (1890s and 1790s) and compare it with the recent years in terms of pollutant fluxes, we chose to use the discharge of 2010, close to a mean hydrological year (see Fig. 2) so as to explore the role played by global changes in sectors such as agricultural practices and land use, industry and waste water collection/treatment, and hydromorphology.

For a true comparison with the present, constraint specificities for historical simulations require knowledge on (i) the point sources as determined from historical archives for industrial and domestic loads, (ii) the diffuse sources from the land use types from old maps and the agricultural nitrogen budget, and (iii) the historical hydromorphology of the river, which has been considerably modified by human activities. Stagnant systems, deeper and wider than the river itself, increase the residence time of the water, subsequently modifying the net growth rate of the biological compartments, and nutrient cycling.

Indeed, human activity has resulted in extensive alteration of the hydraulic regime of rivers beginning in the Middle Ages (10th to 12th centuries). At that time, mills were installed on the smallest streams as well as on the major rivers, and hydraulic energy from water mills played a major role in many activities (Demey, 1990). The construction of embankments along higher-order streams for flood protection also began as early as the 12th century (Décamps et al., 1988). From the middle of the 18th century, development of transport and trade led to

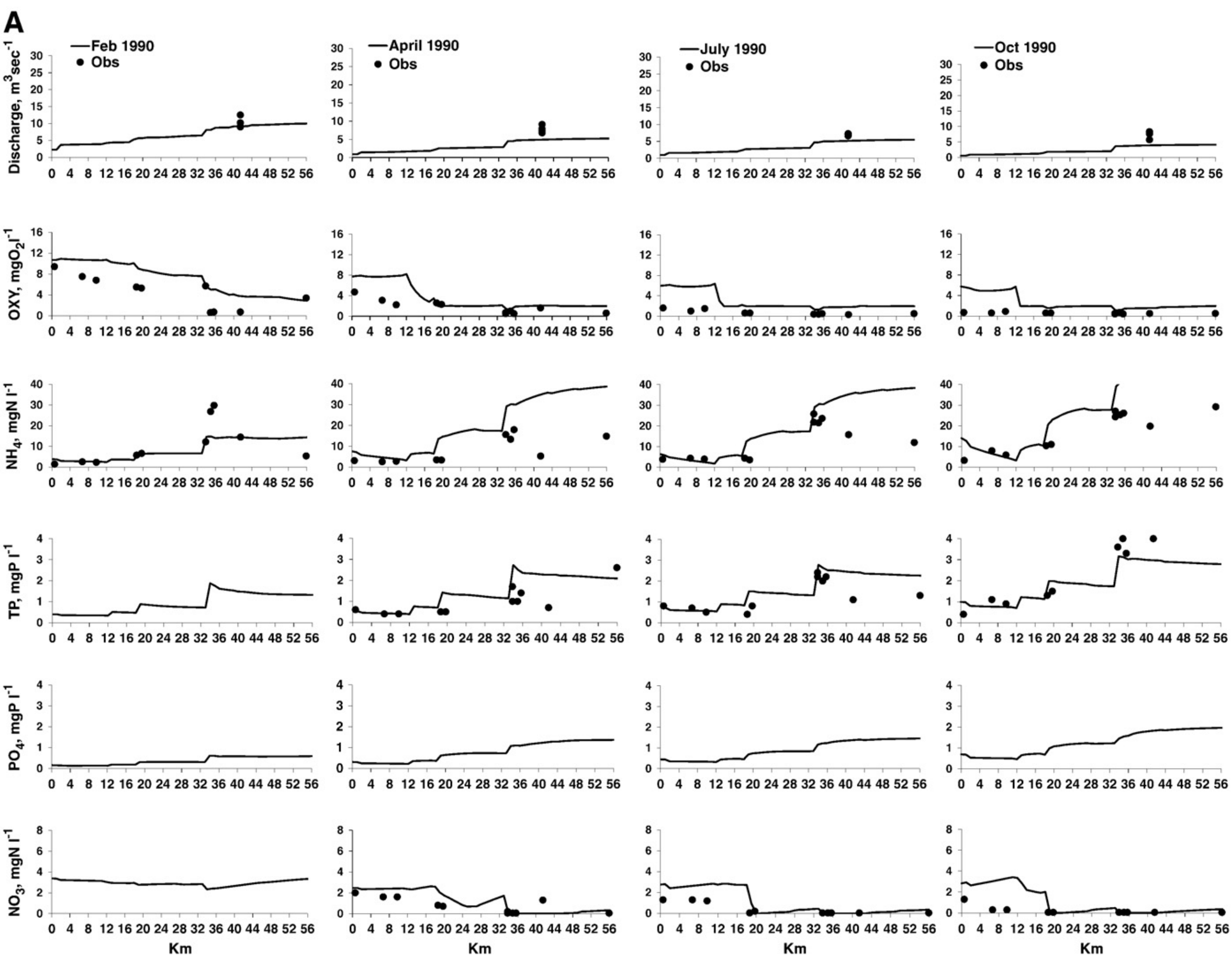

Fig. 4. Longitudinal variations in 1990, 2001 and 2010 (A, B, C), as calculated by the model, of major water quality variables, from Lembeek (km 0) to Heffen (km 56) at the outlet of the river, at four periods in the year (from left to right). From top to bottom discharge ($m^3 s^{-1}$), oxygen (Oxy), ammonium (NH_4–N), total phosphorus (TP), phosphates (PO_4–P), and nitrate (NO_3–N). Observations are shown at Eppegem (km 42) for the discharge and at 10 stations for the other variables. More than one dot indicates other measured values for close to date of the simulations. In 1990, the y-axis is ×2 for NH_4–N compared to 2001 and 2010. In 2010 (D), observations for other variables recently added in the routine survey (silica, phytoplankton expressed as chlorophyll a concentrations, suspended solids and particulate and dissolved organic carbon -POC, DOC-) are also compared with the model results.
Data sources: Vlaamse Milieumaatschappij, Geoloket Waterkwaliteit – http://www.vmm.be/geoview/ – and GESZ research project, Impulse Environment initiative from the Brussels Institute for Research and Innovation.

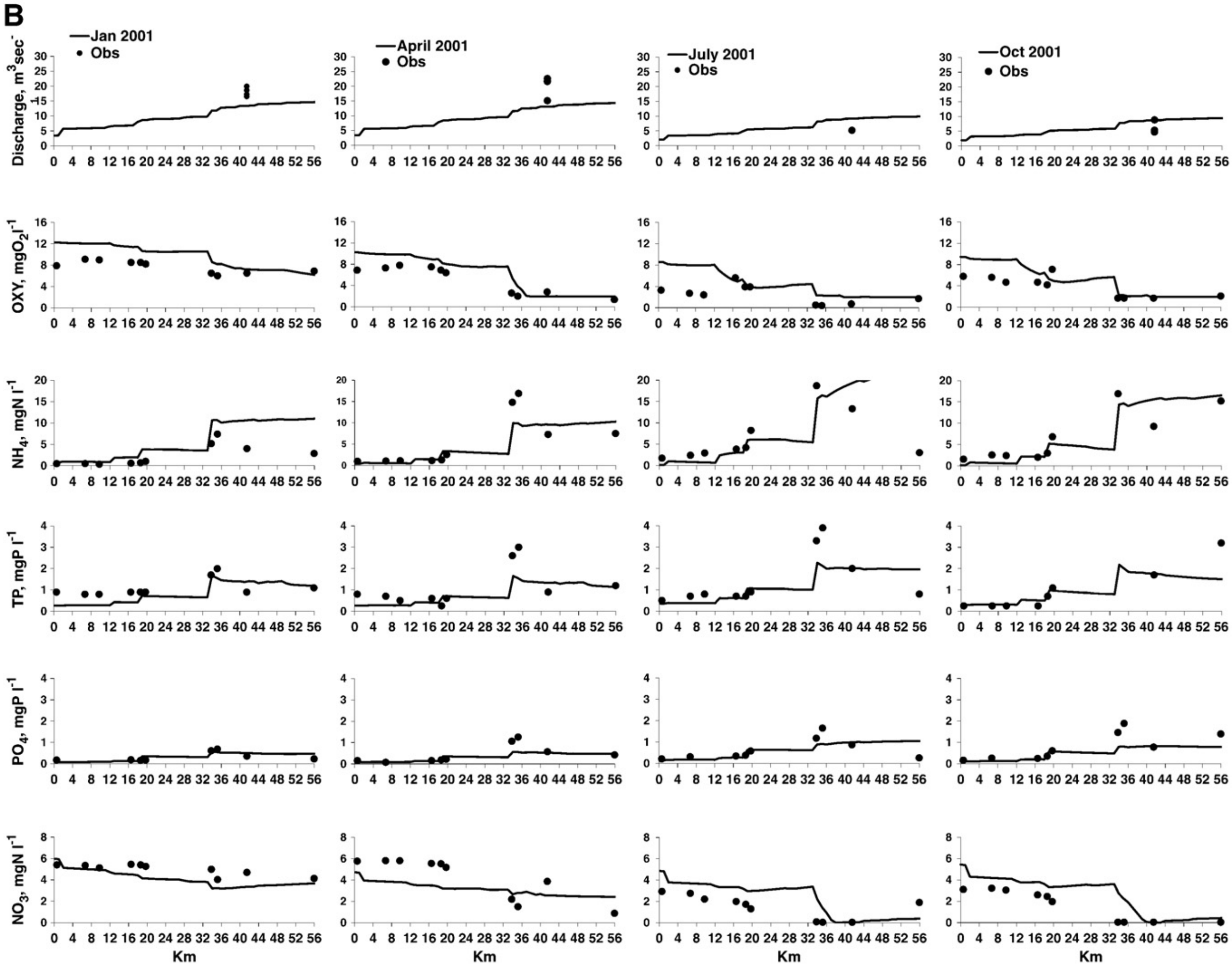

Fig. 4 (*continued*).

large-scale canalization and depth regulation works with construction of locks on the major rivers of Western Europe, and pollution and insanitation of the river due to industrialization led public authorities to cover many small urban rivers in northern Europe, such as the Zenne in Brussels (Deligne, 2003) but also the Bièvre in Paris (Berthier, 2007).

At the same time, many ponds were filled either for public health reasons or because fish breeding or water mill activities ceased after the introduction of the steam engine. In addition, drainage of wetlands, which was widespread in the first half of the 20th century, resulted in complete insulation of the rivers from their flood plain (Amoros and Roux, 1988).

5.1. Hydromorphology

Ponds were inventoried and geo-referenced for the 1890s from the “Dépôt de la Guerre” and “Institut Cartographique Militaire” maps (1865 to 1880, 1:20,000) (hereafter called DLG maps), (Table 4) for the whole Zenne basin. The inventory for the present situation is based on the pan-European urban Atlas (2005), giving information on urban land use for conurbations with more than 100,000 inhabitants (here, Brussels), covering only 67% of the Zenne basin (Table 4). Although no striking differences appear between the two periods, in terms of pond surface area, the shift in their functions, namely from industrial power generation to recreational or ornamental purposes, led to deep changes in their functionality, from closely connected to the river courses in the past to highly disconnected in the present situation, where sand-pit ponds form a large share of the stagnant systems. For these reasons, the results for the present period shown above were considered without taking into account any stagnant annexes in the hydrological network (ponds were not mentioned above for the present times).

To illustrate the long-term historical changes in ponds, we also geo-referenced the map supervised by the Comte de Ferraris (1771–1778, Carte de Cabinet, 1:1152) and inventoried the ponds. The data clearly show a nearly 50% decrease over one century (from the 1780s to the 1880s). As a whole, the surface area of the ponds relative to the surface area of the watershed decreased from 0.36% in the 1790s to 0.21% in the 1890s (see Table 4). Today, despite no change in their surface area, a majority of the ponds are stagnant systems without connectivity with the drainage network and therefore with no influence on the biogeochemical functioning of the river.

Geo-referencing the ponds for the 1790s (Ferraris map) and 1890s (DLG map) at the scale of the Zenne basin and estimating their size provide more information on the historical features. Similar to the Zenne, the extensive alteration of the hydraulic regime of rivers by human activity since the Middle Ages (10th to 12th centuries, Deligne, 2003), is reported for the Paris basin by Guillerme (1990). These stagnant systems were constructed for a variety of functions early in the history of the area (fish ponds: Benoit and Mattéoni, 2004; fish ponds and mills: Deligne, 2003; wood floating for transportation: Benoit and Berthier, 2005) and they were well connected to the rivers, contrary to today's stagnant systems (i) coming from material extraction for construction

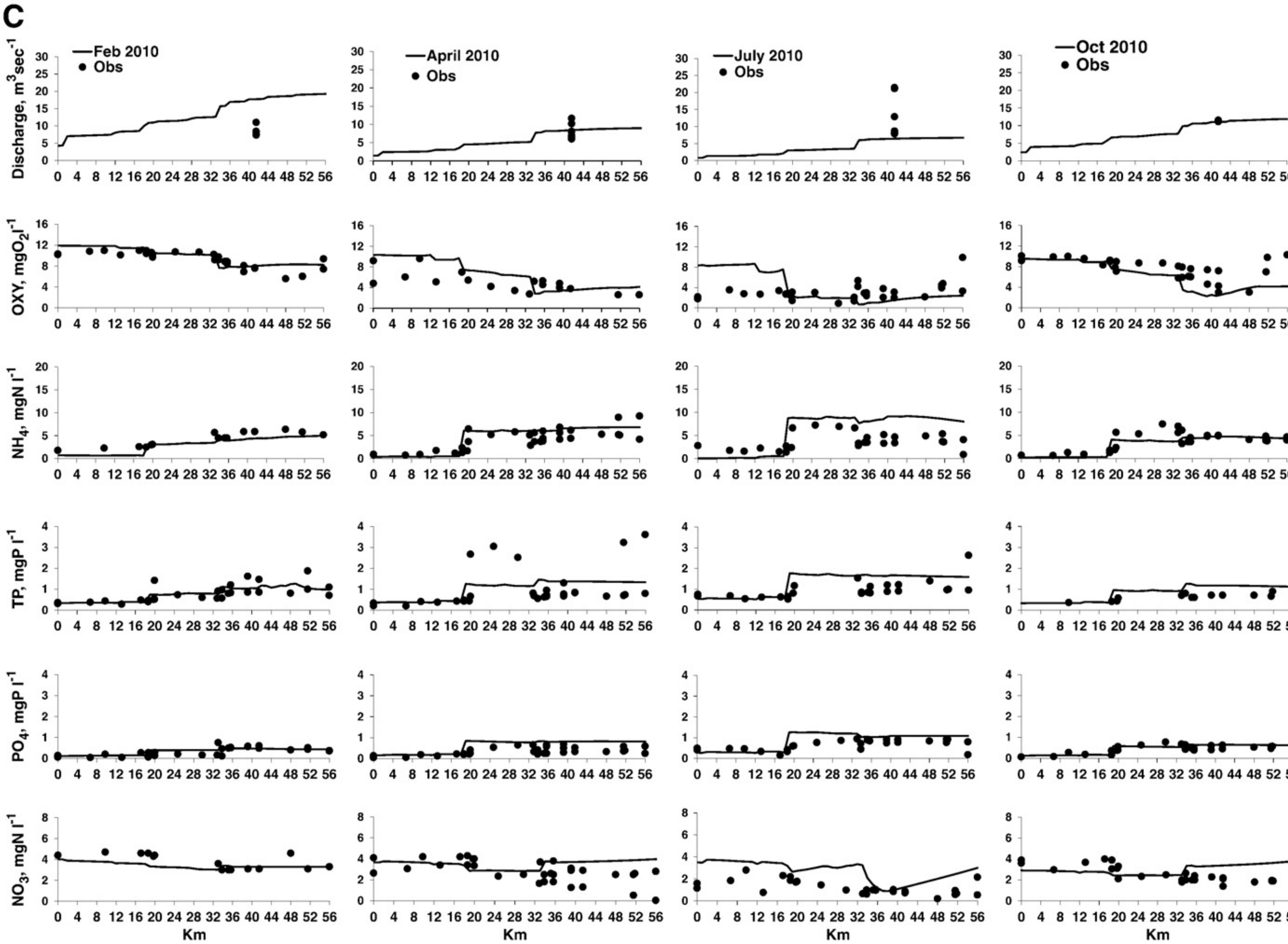

Fig. 4 (*continued*).

(building, roads, etc.), such as sand-pit lakes that were then rehabilitated for recreational use (cf. Davies et al., 2007; Garnier and Billen, 1993; Völker and Kistemann, 2011) or (ii) created in the landscape at the outlet of an agricultural drainage system for fertilizer (nitrate) removal (Passy et al., in revision).

5.2. Domestic point sources

The release of domestic wastewater into surface waters is a characteristic of urbanization. In most traditional agricultural life systems, human as well as animal excrement and domestic wastes are recycled onto croplands (Barles, 2005; 2007). As early as the Middle Ages in Western Europe, however, a primitive system of wastewater collection, often taking advantage of small streams, was organized in even small urban agglomerations for evacuation of domestic wastes to rivers. Modern-type large sewer systems were installed in most large cities in the mid-19th century. Disposal of part of the collected wastewater onto cropland was often organized, but direct discharge into rivers dominated. Physicochemical and biological treatments of wastewater in centralized purification plants began in the early 20th century but became a general practice only after the 1950s.

The "inhabitant-equivalent, Inh. Equ." is the current value of the pollution load corresponding to the mean daily per capita production of domestic wastewater. Stated by decree in most industrialized countries for taxation purposes, in Belgium the regulation defines it as follows (per Inh. Equ.): 90 g day^{-1} suspended solids, 54 g day^{-1} BOD (i.e., 19 g day^{-1} biodegradable organic carbon) and 10 g N day^{-1}. As for N, this value closely corresponds to the physiological level of N excretion (Verbanck et al., 1994). Physiological excretion of P, on the other hand, is evaluated at 1.2 g P day^{-1}, which probably corresponded to the "historical" P Inh. Equ. Note that, it reached 3.5 g P day^{-1} in the 1990s at the maximum use of polyphosphates in washing powders and then decreased to 2 g P day^{-1} from the 2000s when they were banned (see Billen et al., 1999; Garnier et al., 2006; Servais et al., 1999). For silica, the release from domestic wastewater is low with respect to diffuse sources of this element, but not insignificant (Sferratore et al., 2006).

The population data were gathered within the Zenne basin from census statistics (for the 1890s, from the National Statistics of the year 1890; for the 1790s, from the National Statistics of 1800 for Brussels and surrounding towns, and the census from the Napoleonic period in 1801, for the remaining Zenne basin, see Jaumain, 2008). For the 30 cities that were thereby identified for the 1790s and 56 cities for the 1890s, pollution loads were calculated (given the specific pollution load per inhabitant, Inh. Equ.) and were geo-referenced for this study. Whereas the population of the Zenne watershed reached 1.79, 1.82, and 1.91 million inhabitants in 1990, 2000, and 2006, respectively, it was only about 45,000 inhabitants for the 1790s and 300,000 for the 1890s.

We considered that 25% of the waste load was discharged into the Zenne drainage network, exactly as we did for the traditional rural population in the upstream watershed of the Hong-Red River in North Vietnam, where domestic wastewater is not collected, and the river is used for most domestic works, e.g. washing, cleaning, etc. (Le et al., 2010). This assumption seems reasonable because application of domestic sewage as fertilizer onto cropland was still significant at that time (Barles, 2005).

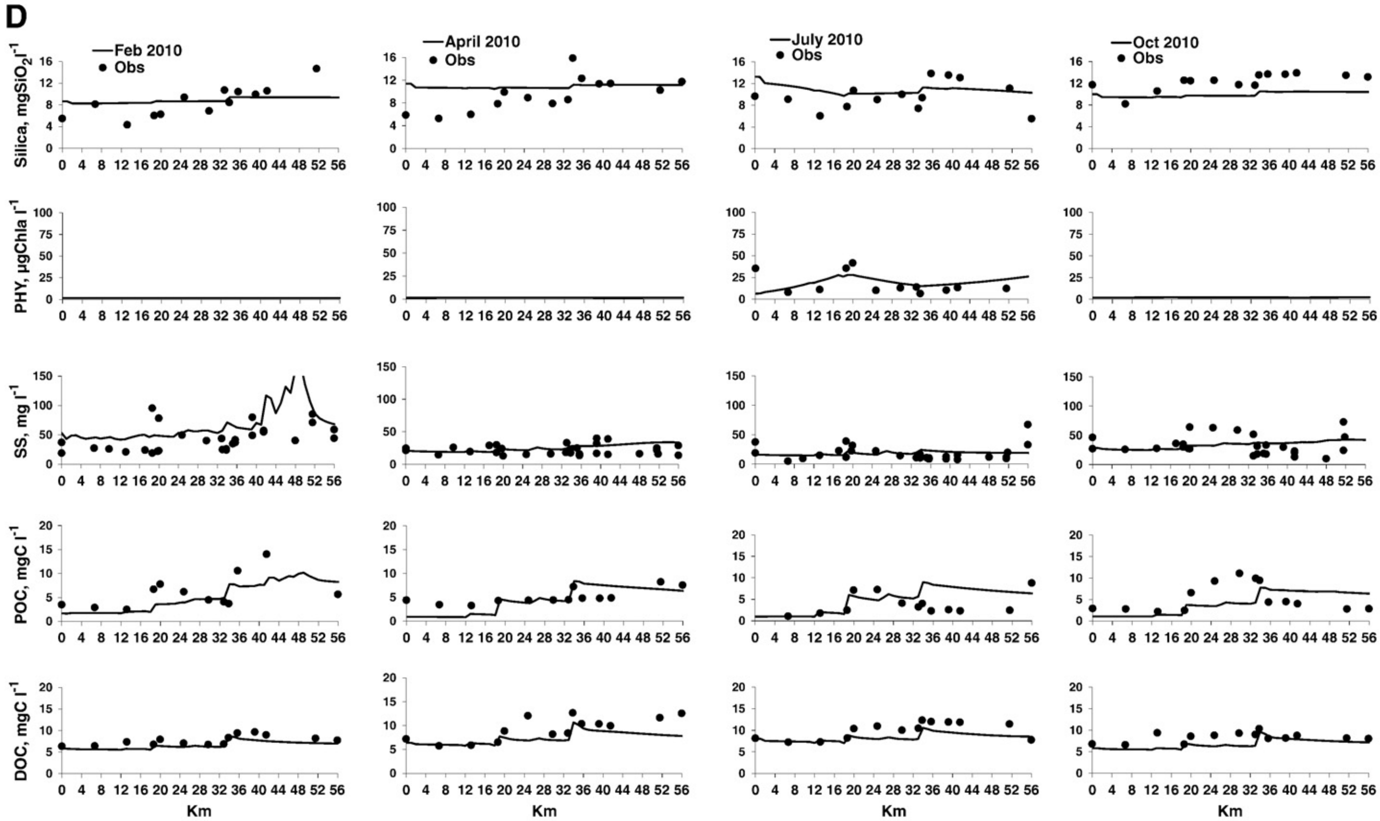

Fig. 4 (*continued*).

5.3. Industrial point sources

Evaluation of the pollution load caused by industrial activities is difficult for historical times. The approach adopted in this study consisted in estimating the industrial load and its spatial distribution from the census of workers by industrial sector (INS, 1876–1900; INS, 1896) and knowledge of the specific pollution load per worker-day for each sector with the technological conditions of historical periods (Billen et al., 1999). These 1896 industrial statistics provide a census of industrial establishments (3751) and associated workers (36,792) distributed among 77 industrial locations, which were all geo-referenced for the present modeling purposes. Ten of these locations situated on the major branch of the river totaled more than 60% of the number of factories and workers. For each of the 12 major sectors known to be a source of pollution (Fig. 5), the loss of organic matter and associated nutrients was estimated from the technical literature of the 18th and 19th centuries (de Beule, 1994; Figuier, 1872; Privat-Deschanel and Faucillon, 1908; Puissant and de Beule, 1989). For many of the activity sectors, the fabrication processes used at an already industrial scale in the 1890s were nearly identical to those in use one century before, on a cottage industry scale. At historical times, however, some reduction of organic and nutrient industrial effluents should have occurred through settlement, because wastewater storage basins, allowing easier management of waste discharges into the rivers, were in common use and became mandatory during the second half of the 19th century (Onclincx, 1991). For the 1890s, the total production per sector and the number of workers for each sector made it possible to calculate the pollution load reaching the river throughout the Zenne basin. Among the various sectors considered, the organic load produced by glue and gelatin and by paper accounted together for 62% of the pollution, and food and beverages (most notably breweries) represented an additional 21%, with the remaining 17% related to a variety of sectors (Fig. 5).

Uncertainties on former industrial processes are difficult to evaluate and depend on the type of process considered. For a given sector the amount of raw material used per unit of final product is documented, so that the organic and nutrient pollution generated can be estimated. For example, Turkey red dyeing illustrates the different steps of the pollution generated: washing with lime; using sheep excrement; oil baths; impregnating with gall; applying alum; dyeing with madder root; boiling with soda, oil and soap; and boiling with tin salts, soap and nitric acid. All these steps also included rinsing and drying phases (Figuier, 1872). The values can be reasonably estimated to vary in a range from 1.5 to 2.

For the 1790s, considering the increase in the population between 1801 and the 1890s, although speculative, we applied a ratio on the same order of magnitude, a factor of 10 for a rough estimation of the industrial load, in order to analyze the ecological status of the Zenne basin, immediately before industrialization became widespread.

5.4. Diffuse sources

A GIS land use layer was established on the basis of the Ferraris map (see above) (Fig. 6). We considered this information representative of the rural landscape over the entire 19th-century period. The most striking difference with the present land use is the obvious extension of urban areas, from 5% to 50%, at the expense of cropland and forest, which decreased from 65% to 25% and from 17% to 8%, respectively. Pasture and orchards were also quite important in the rural landscape at this time, and today they are restricted to scarce riparian sites along the rivers (see Fig. 6).

In order to assess the magnitude of nitrate leaching by arable land in historical periods, a complete nitrogen budget was calculated from agricultural statistics available at the town level for 1880 (Ministère de l'Agriculture, 1885), according to the same procedure as for current budgets (Passy et al., 2013-this issue). The results show that the surplus

Table 4
Changes in the pond surface area (ha) in the various sub-basins of the Zenne River. Ferraris: map of the cabinet (carte de cabinet) 1:1152, supervised by the Comte de Ferraris 1771–1778; DLG: Dépôt de la Guerre map 1:20,000 realized from1865 to 1878 but finished from 1878 to 1880 by the Institut Cartographique Militaire; present is shown for comparison: map from the pan-European urban Atlas, giving information on the urban land use for conurbation > 100,000 inhab, here that for Brussels. The map covers 67% of the whole Zenne basin, but for each of the sub-basin, the documented area varies from 19% and 35% (the Zenne and the Sennette, two upstream watersheds) to 90–100% for all the other sub-basins.

Sub-basins and basin	Watershed area, km^2	Ponds, surface area, ha		
		Ferraris	DLG	Present (67%)
Zenne Axe	192	69	48	97 (87%)
Zenne WSH	236	22	17	5 (19%)
Sennette	272	33	36	24 (35%)
Hain	85	5	4	7 (98%)
Zuunbeek	91	21	11	12 (97%)
Linkebeek	28	13	7	0 (100%)
Pedebeek	22	7	7	10 (100%)
Molenbeek	25	20	5	4 (100%)
Maelbeek	17	50	15	7 (100%)
Woluwe	103	138	55	64 (100%)
Maalbeek	42	28	16	13 (91%)
Kesterbeek	23	4	4	9 (94%)
Barebeek	47	10	12	12 (99%)
Meerbeek	48	20	18	16 (100%)
Total Zenne Basin	1231	440	255	280 (67%)

of N total fertilization over crop N uptake was between 5 and 17 kg N/ha/yr in 1880, while values as high as 80, 250 and 400 kg N/ha/yr were reached in the 1990s in the Loamy, Sand Loamy and Sandy regions, respectively (Table 5). The surpluses then decreased to 50, 175 and 320 kg N/ha/yr in 2006, the last date for which statistics are available and considered here to be representative of 2010.

5.4.1. Modeling water quality in the past

Our modeling framework is a powerful tool to quantitatively assess the overall effects of sometimes opposite historical trends in forcing functions as reconstructed above. We calculated the water quality of the Zenne River with the set of constraints corresponding to the end of the 18th (1790) and the end of the 19th (1890) centuries (Fig. 7). The results show that, among the periods explored, the water quality was the worst in the 1890s, essentially due to the organic and ammonium pollution, typically characterizing the impact of industrial effluents, greater than domestic effluents at these times, contrary to what is observed today.

The results found for the 1790s, despite the speculative assumptions on the industrial loads, show the background water quality of the Zenne River just before the industrial revolution in continental Northern Europe (Belgium and France, followed by Germany) (Fig. 7). Interestingly, after the wastewater purification measures, the present level of pollution returned to a level close to this historical background level in terms of quality.

Typically, the industrial activities of the 1890s were responsible for the pollution of the river more than domestic activities, dating back to the 1850s, taking into consideration the covering of the river in the Brussels district ending in 1871 intended to prevent people from contracting water-borne diseases. Indeed, one century later in the 1990s, because the population had more than doubled, without efficient wastewater treatment, the improvement in water quality corroborates the radical changes in industrial processes at the beginning of the 20th century with the full development of the chemical industry. Such an industrial mutation begun however in the late 18th century (André-Felix, 1971) and chemical products replaced organic products in most industrial processes. In recent times the environmental pollution generated in the Zenne River by the Brussels sewage was hidden in the city due to the rivers' coverage. As a consequence, improving the water quality of the Zenne had not been a priority before the injunction by the European Water Framework Directives (UWWTD, 1991; WFD, 2000).

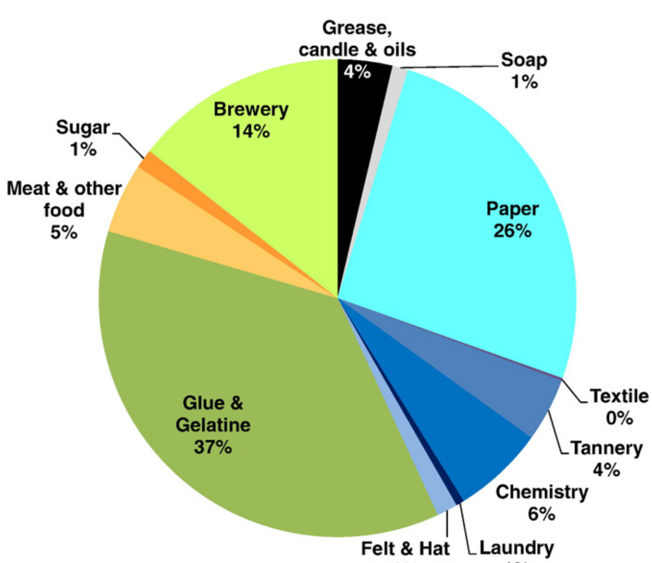

Fig. 5. Distribution of the major sector of industry in 1896 in terms of organic matter load to the river (see text for the data sources and Billen et al., 1999).

6. Role of hydromorphology of the drainage network

For a better understanding of the role of ponds and the covering of the Zenne River in Brussels, seasonal variations of the main impacted variables are shown in Fig. 8 for the downstream station (Eppegem) and for the two historical periods, the 1790s and 1890s, the former being characterized by ten times less pollution, more ponds and the main branch flowing uncovered along its entire course. The present situation (2010) is also shown for comparison.

In order to better understand the role of the ponds, phytoplankton and zooplankton – a major phytoplankton consumer – are analyzed together. Indeed whereas phytoplankton can bloom with a residence time of only a few days in the river, zooplankton requires at least 3 to 4 weeks, a condition found in summer period when the lower discharge and the ponds increase the water residence time in the drainage network. Whereas in both periods phytoplankton reached a higher level than today owing to the presence of connected ponds, especially in spring and late summer, the level was similar for the two historical periods. However, much higher zooplankton biomass at the river's outlet is calculated by the model in the 1790s, supporting the assumption of a much higher phytoplankton production, which was maintained at a similar algal biomass level owing to much higher

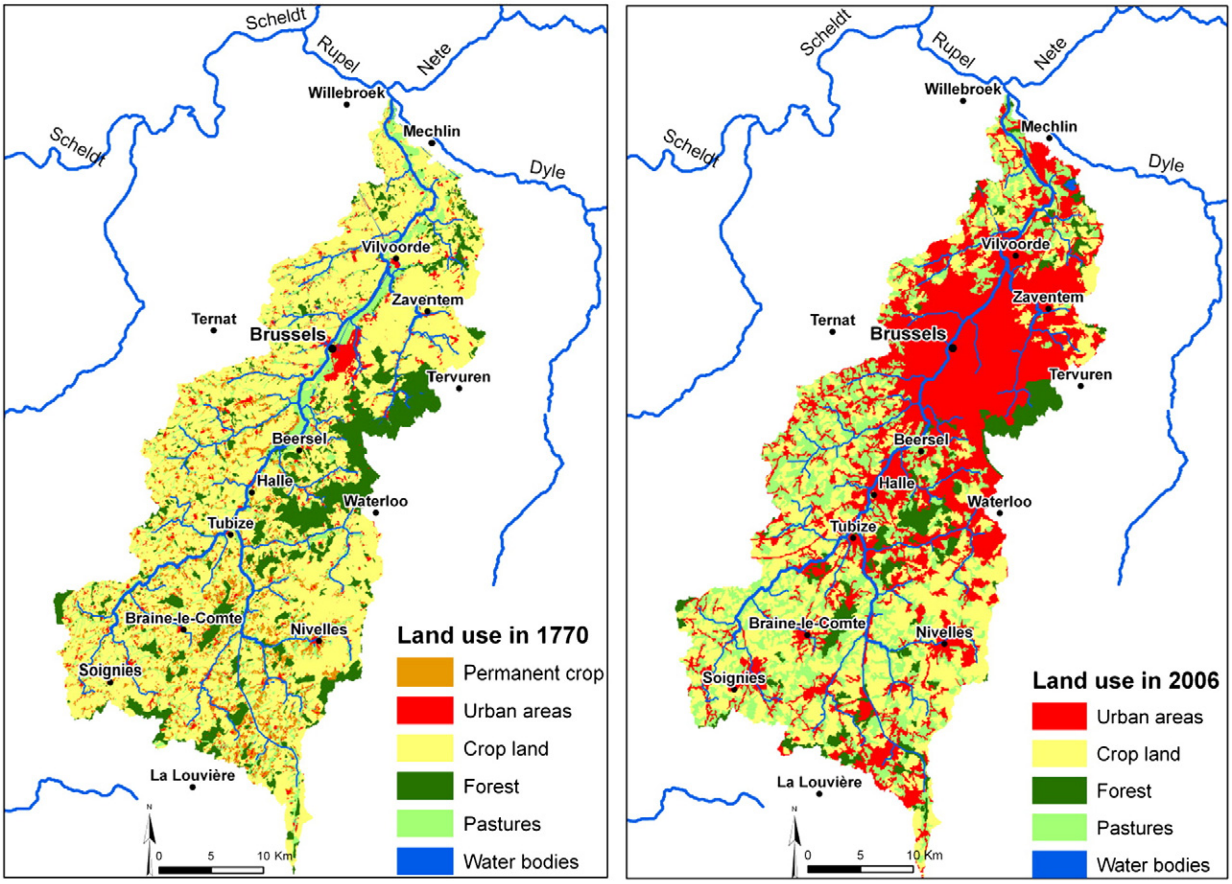

Fig. 6. Extension of the city of Brussels and subsequent changes in land use between Ferraris period and present days (CLC).

grazing in the 1790s than in the 1890s (Fig. 8). Note that it is advantageous to focus on zooplankton here which generally does not grow much in rivers, because the renewal rate of the water is higher than its growth rate (Garnier et al., 1995), except when connected to stagnant systems. The higher silica depletion with ponds, although accentuated in the 1790s, corroborates this interpretation. The higher primary production can be clearly associated with the 20 km of the river receiving light in the 1790s but not in the 1890s (river covered). Therefore, running the model for the two periods with and without ponds brings out the effect of ponds (e.g., residence time) and the effects of covering

Table 5

Long term trends of the nitrogen budget of arable land in the Zenne basin. Data from historical (communal level for 1880) and current agricultural statistics from the Belgian Ministry of Agriculture. Production: agricultural vegetal production expressed in nitrogen unit; LU/ha: Livestock unit per ha agricultural land; Synthetic fert: Synthetic fertilisation; Organic fert: Organic fertilization; Crop N_2 Fix: Crop N2 fixation; Atm depos: Atmospheric deposition; Total fert: Total fertilisation. Surplus = total fertilization − production.

	Year	Production kg N/ha/yr	Livestock LU/ha	Synthetic fert kg N/ha/yr	Organic fert kg N/ha/yr	Crop N_2 fix kg N/ha/yr	Atm depos kg N/ha/yr	Total fert kg N/ha/yr	Surplus kg N/ha/yr
Loamy region	1880	43	0.3	8	17	18.2	5	48	5
	1950	86	0.8	73	47	1.4	5	126	40
	1969	96	1.0	95	58	2.4	10	165	69
	1982	135	1.1	123	87		22	232	97
	1991	137	1.3	123	76	7.6	14	221	84
	1992	133	1.3	123	77	2.8	14	216	83
	1999	169	1.5	126	86	1.7	15	229	60
	2005	204	1.7	128	106	0.6	16	251	47
Sand-loamy region	1880	44	0.4	8	25	14.7	5	53	9
	1950	84	0.9	97	55	3.8	5	161	77
	1969	90	1.5	126	88	2.7	10	226	136
	1982	125	2.3	174	165		21	360	236
	1991	121	3.0	174	178	5.8	14	372	251
	1992	118	3.1	174	182	2.5	14	372	254
	1999	139	3.5	149	209	2.0	15	375	236
	2005	163	4.2	124	198	0.8	16	339	176
Sandy region	1880	50	0.5	8	31	23.8	5	67	17
	1950	82	1.2	128	69	1.5	5	204	122
	1969	85	2.1	166	125	3.6	10	304	220
	1982	114	3.6	226	255		20	501	386
	1991	98	4.7	226	280	6.2	14	527	429
	1992	97	4.8	226	284	4.2	14	529	432
	1999	118	5.3	168	318	3.4	15	505	386
	2005	101	6.2	110	294	1.6	16	422	320

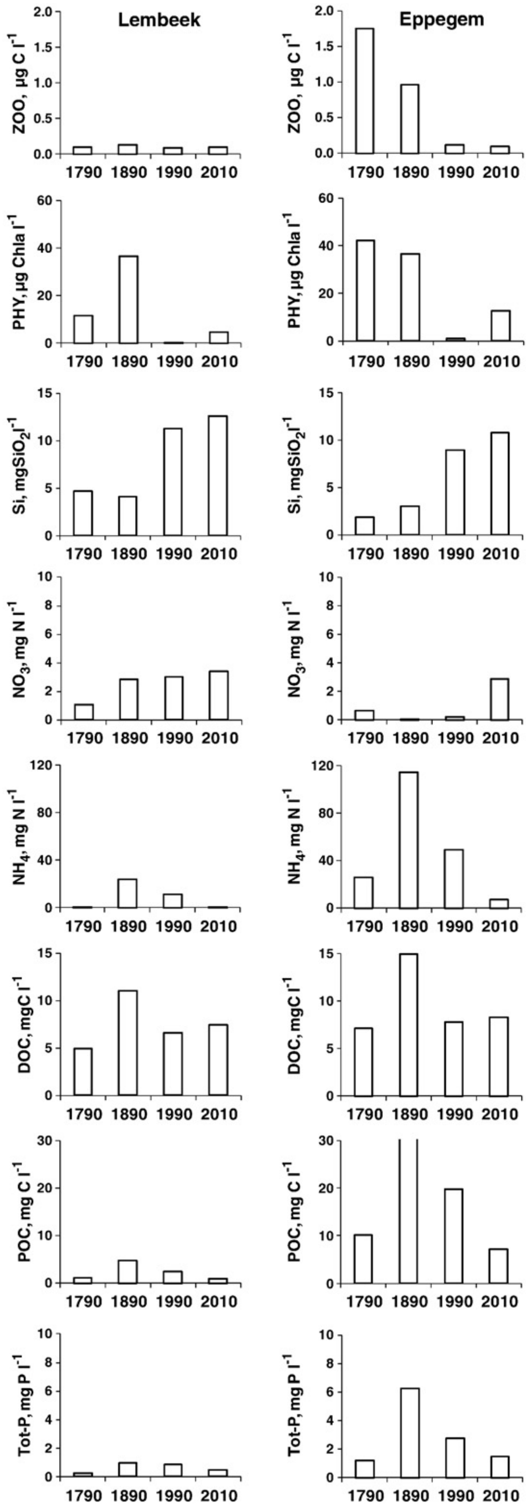

Fig. 7. Evolution of water quality variables for the periods 1790s, 1890s and the years 1990 and 2010 as calculated by the model. The stations at Lembeek and Eppegem, upstream and downstream Brussels on the main branch are represented. ZOO for zooplankton, shown here for its development in 1790 and 1890 with ponds, which increase the residence time of the water.

the river (e.g., light limitation). The presence of ponds, which favored the development of algae and zooplankton, subsequently must have driven freshwater fish production, whose consumption was promoted by the Catholic Church, which imposed lent for 166 days per year (Bérard, 1988; Levasseur, 2005).

Regarding the other nutrients, differences obtained between the two periods for phosphates are linked to the point source constraints, which ranged within a factor of 10, while at high P loads in the 1890s, higher phosphate may be attributed to higher recycling in ponds (Fig. 8). It should again be mentioned that by comparison with the reference in 2010, phosphates were somewhat lower in the 1790s but much higher in the 1890s. Nitrate of diffuse origin logically shows a much higher level today, due to the recent use of synthetic fertilizers in agriculture (Fig. 8). Particularly striking is the disappearance of nitrate in the 1890s, mostly linked to denitrification, a microbial process associated with organic pollution at the origin of anoxic conditions, favorable to the reduction of nitrate into N_2, i.e., elimination into the atmosphere. Denitrification and nitrate elimination are currently observed in aquatic systems where organic pollution is high, even in the 2000s in the Zenne, before the implementation of the Brussels wastewater treatment plants (see Fig. 4), and more generally in the Scheldt estuary up to the 1980 (see Billen et al., 1985 for the 1970s and Soetaert et al., 2006, for a long-term trend).

7. Nutrient fluxes delivered at the basin outlet

In addition to water quality, applying the Seneque/Riverstrahler model also allows one to compare the nutrient deliveries to the estuarine area corresponding to the historical situations explored, assuming identical hydroclimatic conditions (those of 2010) (Table 6). In line with the observations made on water quality, the calculated fluxes delivered at the outlet of the Zenne basin indicate a peak for phosphorus and nitrogen loads in the 1890s and a return in 2010 to the baseline situation simulated for the 1790s. This nutrient trajectory shows the Zenne ecosystem's capacity to recover, after a pollution that probably lasted for two centuries, from the early 1800s to the early 2000s. However, the resilience to nutrient and organic load must differ from that for other pollutants such as metals and persistent organic pollutants (POP: dioxins, PCBs, some pesticides and pharmaceuticals, among others; Meybeck and Helmer, 1989). The behavior of silica is particularly striking, showing a higher flux in 2010 than in the historical periods. Silica is typically trapped in pond sediments, via algal sedimentation (diatoms), thus more in the 1790s than in the 1890s, due to the loss of their surface areas. This higher silica flux for the present period is further accentuated by the fact that the effluents of wastewater treatment plants are also a source of silica; based on per capita consumption of food washing powders and other items containing silica an Inh. Equ. was estimated to 0.45–1.12 g Si capita^{-1} day^{-1} (Sferratore et al., 2006). The median of this range was taken as a point source in the model in the same way as P and N.

In order to characterize the impact of the Zenne River downstream in the Scheldt River, further discharging its nutrient load to the coastal zone of the North Sea, we calculated the ICEP (indicator of coastal eutrophication potential) value of the fluxes discharged at the outlet of the Zenne River (Billen and Garnier, 2007; Garnier et al., 2010). This indicator measures the excess of nitrogen (ICEP-N) and phosphorus (ICEP-P) over silica, expressed in carbon units using the Redfield ratio (Redfield et al., 1963), and leads to the potential for new production of non-diatom algae on the nutrient loading of a river. The indicator values would equal 0 for both N and P if both nutrient loads were in perfect stoichiometry with respect to silica. The results show that the potential for eutrophication by the Zenne contribution to the Scheldt was already high in the past, and that improvements in wastewater treatment were not sufficient to prevent the risk of coastal eutrophication. Indeed, *Phaeocystis* harmful algal blooms have been recorded regularly since 1971 along the Belgian and Dutch coastal zones (Lancelot et al., 1987, 2007, 2011), and mention of their occurrence dating back to the 19th century has been reported (Cadée and Hegeman, 1991; Grossel, 1985).

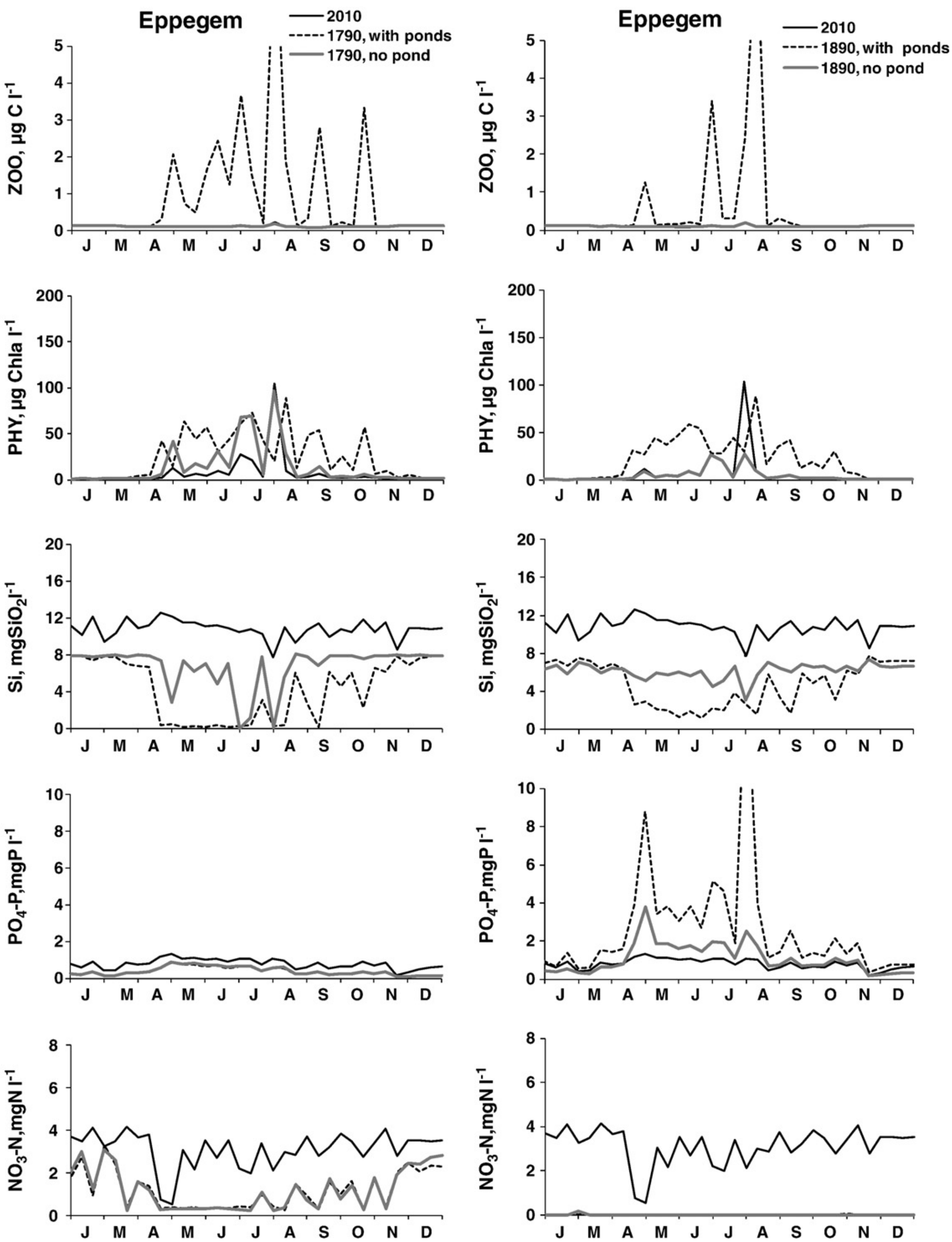

Fig. 8. Seasonal variation of water quality variables as calculated by the model for the 1890s with and without the ponds (ZOO: zooplankton; PHY: phytoplankton; Si: silica; PO_4–P: phosphates; NO_3–N: nitrate). The station at Eppegem downstream Brussels on the main branch is represented. The year 2010 is shown in comparison.

8. Conclusion

Whereas the Seneque/Riverstrahler modeling approach has been used, up to now, for retrospective analysis over a period of time for which experimental data were available (from the 1970s see Billen et al., 2007; 2005; Passy et al., 2013-this issue), this study has shown that the model can be adapted to explore historical periods provided that (i) it has been validated on present well-documented situations and (ii) the past constraint data required can be reconstructed from historical sources. Clearly, this study can be useful in developing countries where familial craft industries are still widespread and for those without efficient wastewater treatments, even in the large cities.

Compared to lake eutrophication (Vollenweider, 1968), the interest in river organic pollution and eutrophication is relatively recent, because rivers have long been seen as systems capable of transporting, purifying and eventually, evacuating downstream all the pollutants discharged into them. Similarly, the link between the coastal eutrophication and nutrient loads from the watershed basins has been reported from the early 1980s only (see Officer and Ryther, 1980 among the earliest papers).

Table 6
Fluxes (Flx) of total nitrogen (N), total phosphorus (P) and silica (Si) as calculated by the model at the outlet of the Zenne River. ICEP-N and ICEP-P, indicators of potential eutrophication, represent a risk of promoting undesirable algae when positive (e.g. N, P in large excess to silica).

	FlxN kg N km^{-2} day^{-1}	FlxP kg C km^{-2} day^{-1}	FlxSi kg P km^{-2} day^{-1}	ICEP-N kg Si km^{-2} day^{-1}	ICEP-P kg C km^{-2} day^{-1}
2010	8.0	0.8	3.8	36.8	25.9
1890s	50.6	2.3	1.5	284.0	89.9
1790s	6.2	0.3	0.9	33.3	9.1

The results of the detailed investigations carried out within the scope of this project in close collaboration between historians, biogeochemists and ecologists offer new insight into the past trends of nutrient delivery at the outlet of an urban basin. It reveals the importance of industrial sources of organic matter and nutrient contamination of surface waters by industrial activities as early as the middle of the 19th century. Industrial pollution preceded, rather than followed, domestic pollution in West European countries. Our analysis also revealed that, owing to this substantial and early pollution by the industrial sector, point discharge of nutrients into surface waters may have been maximum at the turn of the 19th century, leveling off long before the EU-WFD reversed the trend.

Acknowledgments

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