

Assessing the impacts of wastewater treatment implementation on the water quality of a small urban river over the past 40 years

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Abstract This paper studies the effects of the implementation of wastewater treatment (WWT) on the water quality of small urban river systems by considering as an extreme case study (volumetric contribution of wastewaters >50 %) the evolution of the Zenne River waters (Belgium) over the last 40 years. In urban rivers, organic matter (OM), oxygen, and nutrients are primarily controlled by wastewater releases which depend on the population and the WWT capacity in the river basin, the latter being dependent on environmental policy decisions. We introduce a novel basin-scale evaluation method that considers the evolution of annual pollutant loads at the outlet of the river basin directly as a function of WWT capacity. Based on this approach, we could prove that the load reductions observed after the implementation of WWT in the river basin was a good indicator of the global treatment efficiency of the WWT plants. We also show that high self-purification processes within the river basin may lead to reach minimum levels of OM before the completion of WWT. In addition, the effects of wet weather conditions did also change as a function of the WWT capacity going from positive effects at low capacity to negative effects at high capacity. Finally, the

full implementation of WWT in urban river basins does not necessarily guarantee a good status for water quality, mostly because of the high volumetric proportion of treated wastewaters, which do not have the quality standards of river waters.

Keywords Urban river · Wastewater treatment · Treatment efficiency · Long-term trends · Outcome of Directive 91/271/EEC · Oxygen · Nutrients · Organic matter

Introduction

Since historical times, numerous large cities, with their very high population densities, have been developing in the vicinity of water bodies (lakes, rivers) to provide sufficient freshwater resources. These water resources are not only essential to ensure human's vital functions, they are also important as transport routes and for the production of their food, goods, and energy. Moreover, freshwater bodies close to the cities, and especially flowing rivers, are also often used as evacuation routes for all kinds of waters polluted by domestic and industrial waste products. It is well known that this results in the contamination of these waters by pathogenic microorganisms (bacteria, viruses, protozoa) and by a variety of chemical substances that can be more or less harmful for the environment and/or the human health. Among these substances, urban waters are especially rich in suspended solids, organic matter, and mineral nutrients, such as nitrogen and phosphorus (see for example Servais et al. 1999) originating from domestic activities, and from industries (especially food industries). Increased inputs of organic matter and mineral nutrients to rivers have an ecological impact—or footprint. Indeed, they increase the oxygen consumption through the mineralization of the organic material and increase the risk of eutrophication in the river.

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For a given populated zone (for example a city), Rees et al. (1996) defined the “ecological footprint” as the area of productive land and water ecosystems required to produce the resources consumed by that population and assimilate the wastes that this population produces. If we apply this concept to an urban zone located along an aquatic ecosystem and focus on waste evacuation, we could say that the ecological impact of an urban area towards its river is defined as the river flow required for assimilating the wastewater loads that are generated by the city. Hence, the ecological impact of a city towards the river will depend on (1) the river water flow which defines the dilution capacity and depends on the watershed size, morphology and physiography, and on the rainfall; (2) the urban wastewater loads to be assimilated which primarily depend on the population size but can be reduced artificially by wastewater treatment; and (3) the natural self-cleaning potential of the receiving river.

The potential threat of the wastewater load of any city towards its receiving river can be evaluated by considering the “population stress” defined as the ratio between population and average river discharge (Meybeck 1998). A comparison between a selection of different European cities based on this criterion highlights a large variability (more than three orders of magnitude, Fig. 1). It is hereby noticed that big cities sometimes discharge in small rivers (e.g., the 3.26 M inhabitants of Madrid discharge in the Manzanares River that has an average discharge of $1 \text{ m}^3 \text{ s}^{-1}$) and smaller cities may discharge in large rivers (e.g., 1.73 M inhabitants of Vienna discharge in the Danube that has an average discharge of $1915 \text{ m}^3 \text{ s}^{-1}$). On the other hand, cities as different in size as Brussels (1.13 M inhabitants) and Moscow (11.5 M inhabitants) are similar when compared to the discharge of their receiving rivers (around 300,000 inhabitants per $\text{m}^3 \text{ s}^{-1}$).

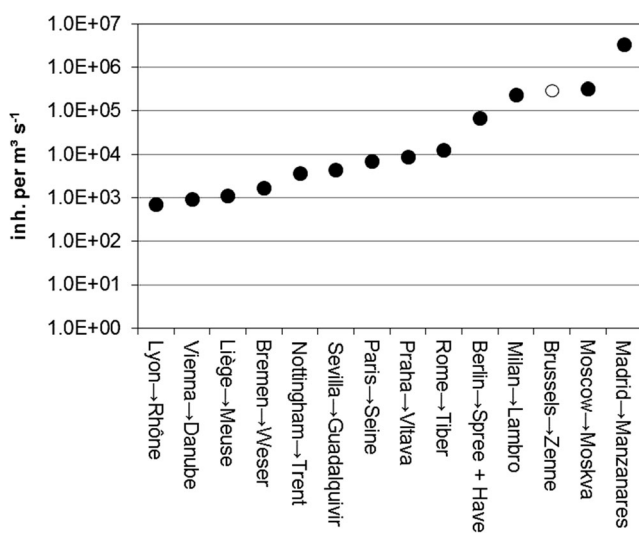


Fig. 1 Population stress (ratio between population of the city and average annual discharge of the river upstream the city) on various rivers draining some European cities; log scale; white dot indicates Brussels

The artificial reduction of wastewater loads depends on the wastewater treatment capacity and technology of the cities. Indeed, the purpose of wastewater treatment plants is to decrease the suspended matter (primary treatment), organic matter (secondary treatment), and nitrogen and phosphorus (tertiary treatment) content of the sewage before its release to the river, and this strongly mitigates the negative environmental effects of the urban effluents.

In the European Union, the Directive 91/271/EEC required at least a secondary treatment of the sewage waters for all cities of more than 15,000 inhabitant equivalents (IE) before the year 2000, and by 2005 for all the cities of more than 2000 IE. In addition, in zones classified as “sensitive” to eutrophication, a tertiary treatment is now required for cities with more than 10,000 IE. As a consequence, this accelerated the implementation of wastewater treatment plants in European river basins over the last 25 years, with positive feedbacks. Surprisingly, only a few experimental studies reported the direct consequences of these changes on river water quality evolution.

The general objective of this paper is to assess the efficiency of reducing wastewater emissions on the water quality of a small urban river. For this, we need to understand the respective effects of (1) wastewater treatment management changes, (2) natural meteorological variability, and (3) in-stream retention processes on the water quality. Among all European cities, the impact of Brussels on its receiving water (the Zenne River) is among the most extreme (Fig. 1) and will be considered as a case study by considering the evolution of organic matter, oxygen, and nutrient (N and P) concentrations over a long time period (40 years) covering minimum to (almost) full treatment of domestic wastewaters emitted in the river basin.

Material and methods

Site description

The Zenne River is a relatively small river with an average annual discharge of $3.7 \text{ m}^3 \text{ s}^{-1}$ upstream from Brussels and $10.0 \text{ m}^3 \text{ s}^{-1}$ at the outlet of the basin (deduced from water flow data of the last 25 years as monitored by the Hydrologisch Informatie Centrum of the Flemish Region—HIC). It is located in the Scheldt River basin and is a tributary of the Dijle River. The Zenne River basin covers an area of 1160 km^2 , with important agricultural activity in the upstream part (51 % of arable land and 18 % of pasture land), a high urbanization (19 %), and a minor presence of forests (10 %) (Shrestha et al. 2013). The Zenne watershed is characterized by a very high population density (average in 2013 1480 inh km^{-2}) especially in the Brussels conurbation (7057 inh km^{-2}). The river’s hydro-morphological characteristics have been strongly modified over the course of the past

centuries. The most important changes concern the diversion of part of the upstream base-flow to feed a navigation canal (Charleroi-Brussels canal), the use of this canal as a bypass during flood events and the vaulting of a 7-km-long river stretch in the center of Brussels city. Presently, major human perturbations on the river are sewage disposals in the Brussels region, as documented by the present study and overall pollutant budgets performed earlier (Vanderborght et al. 2007). The Zenne River crosses Brussels from south to north, and, on average, the sewage disposals add $3.8 \text{ m}^3 \text{ s}^{-1}$ of wastewaters to the $3.7 \text{ m}^3 \text{ s}^{-1}$ of water arriving in Brussels from the upstream part of the watershed. Hence, on average, more than half of the water volume carried by the river downstream from Brussels is sewage, this proportion being even higher during the low-flow periods. The implementation of wastewater treatment in the Zenne River basin occurred stepwise in time and started relatively late. Until 1986, only few wastewater treatment plants (WWTPs) were operating and the treatment capacity did not exceed 10,000 IE for the whole river basin (total population 1.7 M inhabitants in 2013). A few larger plants were built in the 1990s, mainly upstream from Brussels, but it is only after 2000, under the pressure of European legislation, that a significant proportion of the wastewater was treated. Only after 2007 did this practice become general (Fig. 2).

Finally, the hydro-climatic conditions in the Scheldt River basin are mild and maritime with fluctuating precipitation patterns and, hence, river discharge.

The considered monitoring locations are within the main river stream of the Zenne that flows over ± 50 km between Lembeek (reference station for measuring distances—0 km) and the confluence with the Dijle River (Fig. 3). Within this stretch, the river receives the minor water contribution from small tributaries from which the largest ones are the Zuunbeek (19 km) and the Woluwe River (35 km) (respective average discharge of 0.6 and $0.15 \text{ m}^3 \text{ s}^{-1}$). The city of Brussels extends from 20 to 34 km, and the most central portion of the river, a stretch of about 7 km, has been covered during the nineteenth

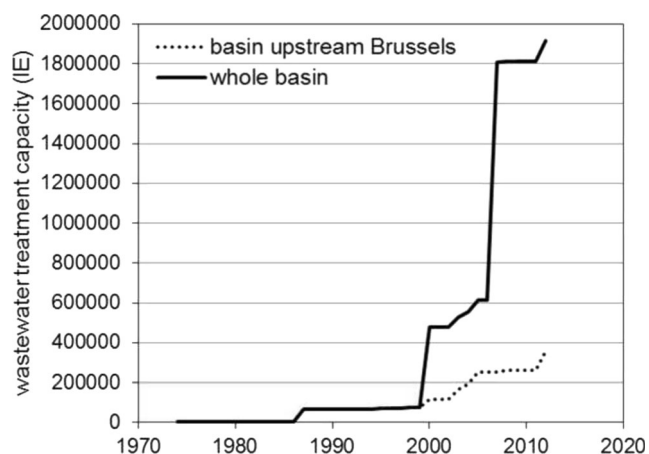


Fig. 2 The total wastewater treatment capacity (IE) in the Zenne River basin upstream Brussels and in the whole river basin from 1974 to 2013

century (1867–1871) for sanitation purposes. The wastewater from the city is collected by a combined sewer system and is presently treated by two WWTPs: Brussels South WWTP with a capacity of 360,000 IE and secondary treatment technologies (no specific removal of N and P) since the year 2000 and Brussels North WWTP with a capacity of 1,100,000 IE and tertiary treatment technology (simultaneous removal of C, N, and P in an activated sludge process) since the year 2007. During high-discharge wet weather conditions, both WWTPs also rely on a parallel “Rain” line treatment procedure based on primary settling processes, coagulation-enhanced (Actiflo®, in North WWTP) or not (South WWTP).

Parallel to the river, the navigation canal is also used as a storage buffer for extreme flow events. There are three overflow structures from the river to the canal to mitigate discharge peaks and protect the city from flooding (upstream from Brussels at 1.2 km and inside Brussels at 20.1 and 29.7 km; Fig. 3), and one overflow from the canal to the river to restore the water budget of the river downstream from the city (39.6 km, Vilvoorde).

Data collection and treatment

Water quality data from 1990 to 2013 were obtained from the Flemish environmental agency (Vlaamse Milieu Maatschappij—VMM) through their online data portal “Geoview” (<http://geoloket.vmm.be/Geoviews/>; 29/09/2014). Data from the 1970s are from reports of the Waterbouwkundig Laboratorium (1977, 1980). Data from the 1980s are from the reports of the Instituut voor Hygiene en Epidemiologie (1979, 1982, 1984, 1985, 1986, 1987, 1988, 1989) and from Smet (1990).

Considered variables are temperature, dissolved oxygen (O_2 , concentration and % saturation), biological oxygen demand (BOD), chemical oxygen demand (COD), N-ammonium (N-NH_4), N-nitrate (N-NO_3), total nitrogen ($\text{Ntot} = \text{sum of Kjeldhal nitrogen and N-NO}_2 \text{ and N-NO}_3$), P-phosphate (P-PO_4), and total phosphorus (Ptot). Measurements are reported to be performed following the standard methods for water and wastewater analyses.

From the available set of monitoring locations, four stations were selected: Lembeek (0 km) and Drogenbos (18.6 km) which are located upstream from Brussels, and Vilvoorde (35.15 km) and Eppegem (41.5 km) which are located downstream from Brussels (Fig. 3). In addition, we also considered stations located at the outlet of the Zuunbeek (St Pieters-Leeuw) and the Woluwe (Vilvoorde) rivers.

Water discharge data are available as daily averages and were collected for the stations “Lot,” “Vilvoorde Sluisstraat,” and “Eppegem” (Fig. 3) at the dates corresponding to the water quality data (in order to be directly compared to concentration data and calculate loads). For the period after 1980, data were obtained from the HIC (Waterbouwkundig

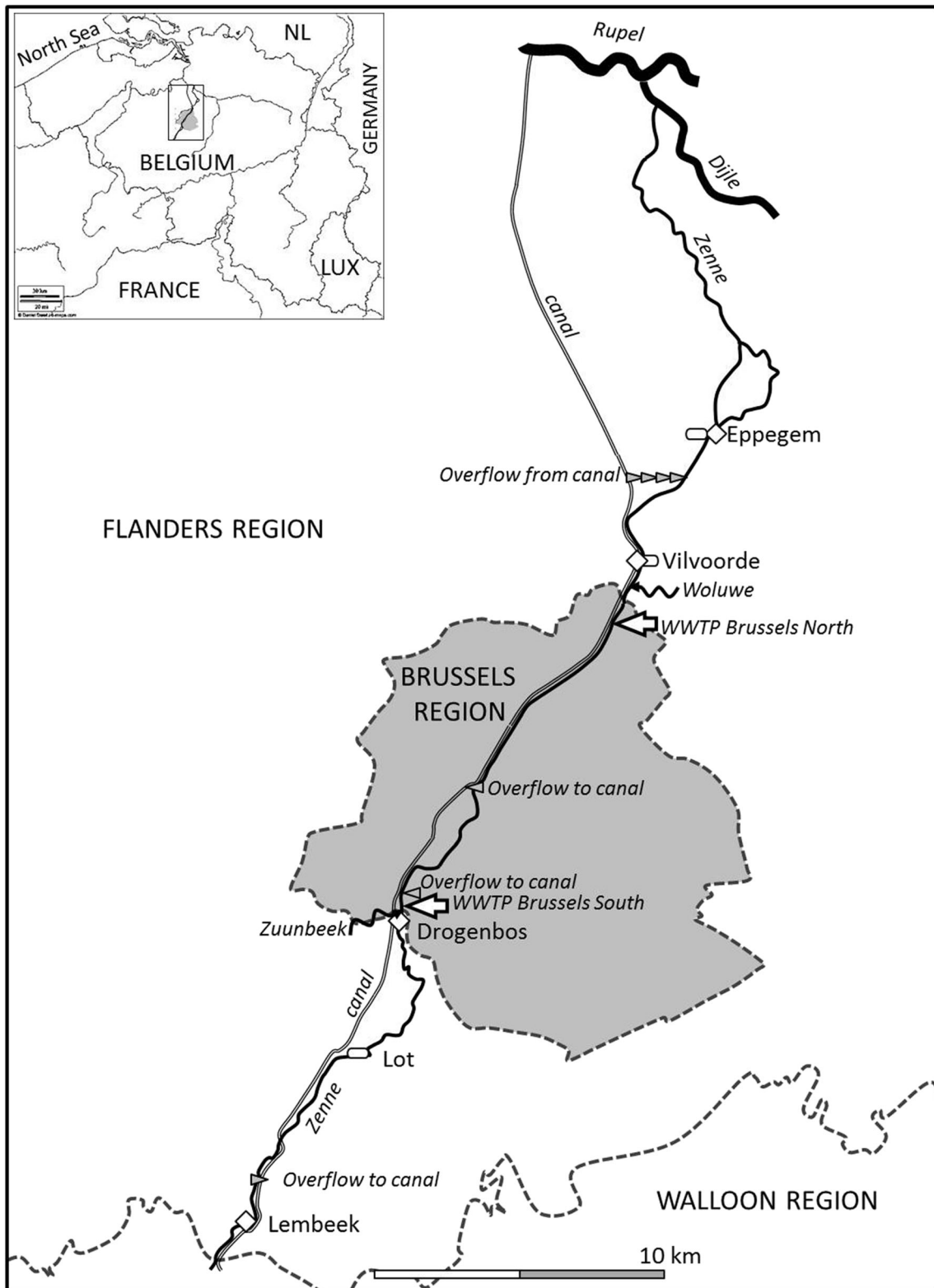


Fig. 3 The Zenne River between Lembeek and the confluence with the river Dijle. Indicated are the location of the Brussels WWTPs (white arrows), the outlet of major tributaries Woluwe and Zuunbeek rivers (wave-shaped arrows), the VMM water quality monitoring stations

(diamonds), and the HIC water discharge monitoring stations (elliptic shapes). Dotted lines indicate the limits of the administrative Brussels, Flanders, and Walloon regions

Laboratorium, Berchemlei 115, B-2140 Antwerpen, Belgium) of the Flemish region authorities as water depths (1980–1989) that were processed into discharges by the application of the depth-discharge rating curves applicable at the time, and as direct discharge values (1990–2013). Data from the 1970s are from reports of the Waterbouwkundig Laboratorium (1977, 1980). In addition, we also used hydrometric data for the Woluwe and the Vogelzangbeek (a tributary of the Zuunbeek) from Flowbru online records (<http://www.flowbru.be/fr/>; 22/12/2014), and for Zuunbeek from VMM through the data portal “Water Info” (<http://www.waterinfo.be/>; 22/12/2014).

For the water quality data, the number of observations per year strongly varies in function of the monitored variable and the observation period. In addition, sampling dates and frequency also vary according to station. In the 1970s and 1980s, there were on average 4 observations per year and per station (one per season), while after 1990, there were between 12 and 50 observations per station and per year. In order to have sufficient data for the analyses, the observations for the years 1970s and the years 1980s were grouped per decade and considered as the 1970s and the 1980s periods. The latter grouping is considered acceptable, as nearly no wastewater treatment was performed during this period and therefore similar conditions prevailed with respect to the wastewater input. For the period after 1990—when a sufficient amount of observations is available and when the WWTP infrastructure was progressively implemented—a detailed year-to-year variability was considered.

For all variables, yearly averages were calculated and significant correlations were sought using the Spearman rank order correlation coefficient.

Results

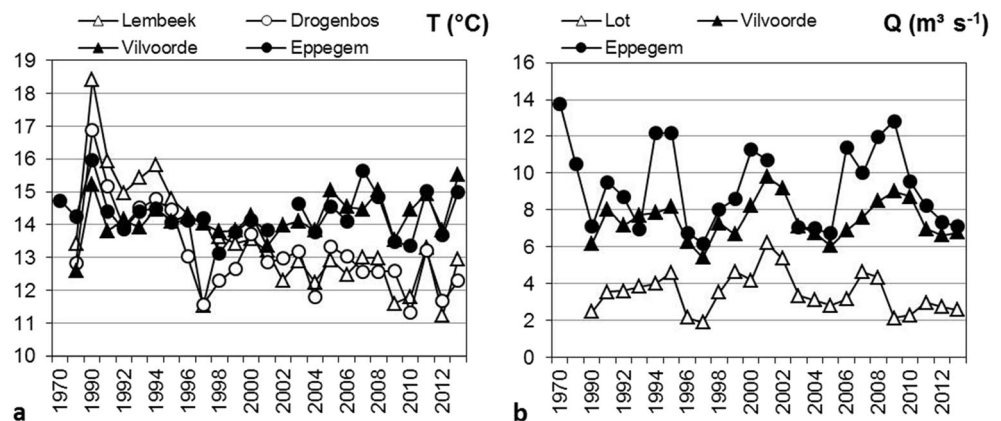
Hydro-meteorological variables: water temperature and discharge

Figure 4 shows that there is a predominant effect of Brussels on the water discharge. As presented before, the sewage

disposals of the city add on average a discharge of $3.9 \text{ m}^3 \text{ s}^{-1}$ of wastewaters to the river flow. There are also substantial year-to-year differences in the discharge distributions at all stations (Fig. 4). The years 1996–1997, 2003–2005, and 2010–2011 were relatively drier (lower discharge—average 2.6 and $7.0 \text{ m}^3 \text{ s}^{-1}$ for the up- and downstream stations, respectively), while the years 2000–2002 and 2006–2009 were relatively wetter (higher discharge—average 4.2 and $10.2 \text{ m}^3 \text{ s}^{-1}$ for the up- and downstream stations, respectively). Finally, the discharge at Lot, Vilvoorde, and Eppegem are following similar trends, except a few cases. For example, in 1994, 1995, and 2006, high annual discharges are observed in Eppegem while not in Lot nor in Vilvoorde. This discrepancy is attributed to a much lower number of sampling days these years in Eppegem (8 to 10 per year) compared to those in Lot and Vilvoorde (22 to 52 per year), and an important flood event on one of the dates triggering averages towards higher levels in smaller datasets. Temperatures also display interesting fluctuations. Upstream from Brussels, the temperatures decreased from the early 1990s (average $15 \text{ }^\circ\text{C}$) and were stable after 2000 (average $13 \text{ }^\circ\text{C}$). As no similar trend was observed downstream from Brussels (Fig. 4), the observed decrease upstream is probably not related to meteorological variability but rather to the progressive deindustrialization of the region upstream from Brussels, resulting in less warm industrial water discharges. After 2000, the temperatures downstream Brussels are higher as compared to those upstream (by about $2 \text{ }^\circ\text{C}$) probably because of the decreased upstream temperature and the large contribution of warmer treated sewage.

It is important to consider that the average annual river discharge that is presented here, as calculated based on a limited number of selected sampling days, may not necessarily reflect the true average conditions over the whole year. This was checked by comparing our values with the annual averages calculated using all days in a year. It appears that the calculated yearly average on selected sampling days varied between 77 and 110 % (P10% and P90%, respectively) and is on average 94 % of the true yearly mean. We can thus

Fig. 4 Evolution from the 1970s to 2013 of **a** water temperature (T) and **b** average annual discharge (Q) in Lembeek or Lot, Drogenbos, Vilvoorde, and Eppegem. Open symbols denote upstream from Brussels, and black symbols downstream from Brussels



consider that they are reasonably representative of the observation year.

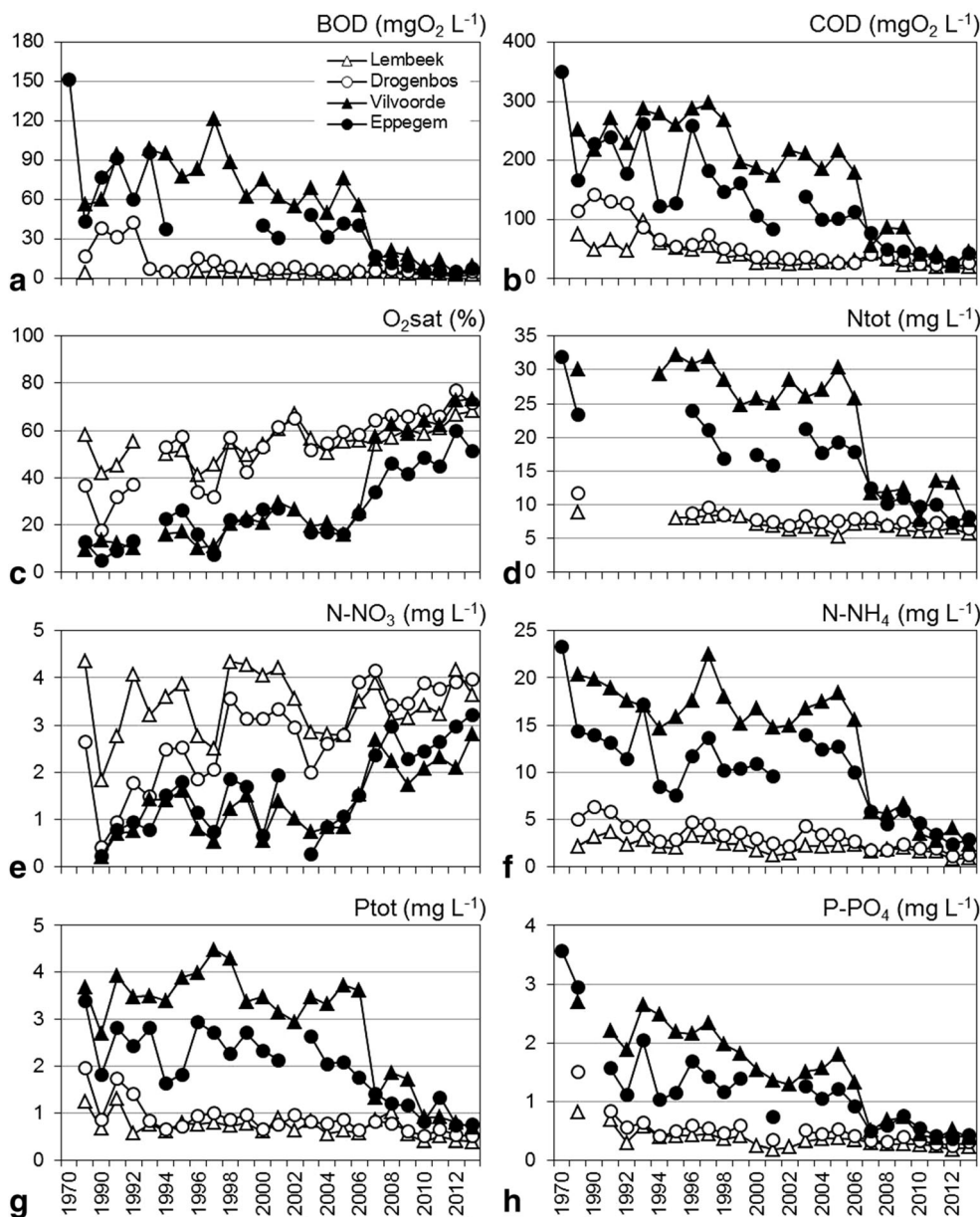
Organic matter and oxygen (BOD, COD, O₂)

As for the water discharge, there is a predominant effect of Brussels on the organic matter as can be seen by the difference in yearly average concentrations between Drogenbos and Vilvoorde (+12 to +69 mg O₂L⁻¹ for BOD and +55 to +193 mg O₂L⁻¹ for COD, P25% and P75%, respectively) and oxygen saturation levels (-5 to -33 % saturation, P25% and P75%, respectively) in the river (Fig. 5a–c).

Total and biodegradable organic matter, as assessed by COD and BOD measurements respectively, decreased at all stations over the studied period (Fig. 5a, b). Upstream from

Brussels, this started in the early 1990s (1992–1993, on average for the two upstream stations 24 and 92 mg O₂L⁻¹ for BOD and COD, respectively) and stabilized around the year 2000 (on average 5 and 28 mg O₂L⁻¹ for BOD and COD, respectively). Downstream from Brussels, the decrease occurred more stepwise: a first small decrease after 2000 (on average for the two downstream stations, from 80 to 50 mg O₂L⁻¹ for BOD and from 228 to 154 mg O₂L⁻¹ for COD) and a second, more pronounced, one after 2006 (on average from 50 to 12 mg O₂L⁻¹ for BOD and from 154 to 55 mg O₂L⁻¹ for COD). Similarly, oxygen saturation levels increased everywhere (Fig. 5c): upstream from Brussels (especially in Drogenbos), we notice a gradual increase since the early 1990s (on average, from 40 to 60 %), while a more stepwise increase is observed downstream from Brussels:

Fig. 5 Evolution from the 1970s to 2013 of the average annual **a** biological oxygen demand (BOD), **b** chemical oxygen demand (COD), **c** dissolved oxygen saturation (O₂sat), **d** total nitrogen (Ntot), **e** ammonium (N-NH₄), **f** nitrate (N-NO₃), **g** total phosphorus (Ptot), and **h** phosphate (P-PO₄) in Lembeek, Drogenbos, Vilvoorde, and Eppegem. Open symbols denote upstream from Brussels, and black symbols downstream from Brussels



from an average of 15 % before 2000 to 22 % between 2000 and 2006 and to 55 % after 2006.

Total nitrogen, ammonium, and nitrate

As for the other variables, there is a predominant effect of Brussels on the nitrogen content of the river water (+6 to +22 mg L⁻¹ of N_{tot}, +4 to +13 mg L⁻¹ of N-NH₄, and -1.1 to -1.8 mg L⁻¹ of N-NO₃, P25% and P75%, respectively, between Drogenbos and Vilvoorde) (Fig. 5d–f).

Although there is a lack of data in the early 1990s, the total nitrogen clearly decreased upstream from Brussels (Fig. 5d) (on average from 10 to 7 mg L⁻¹) until the year 2000 and stabilized afterwards. Downstream from Brussels, as already seen for the other variables, the decrease occurred stepwise and is especially important between 2006 and 2007 (on average from 27 to 11 mg L⁻¹). Similarly, upstream from Brussels, N-ammonium gradually decreased (Fig. 5e) between the 1980s and the year 2000 (on average from 4 to 2.2 mg L⁻¹). Downstream, a sharp decrease was observed after 2006 (on average from 14 to 5 mg L⁻¹). Finally, it is noticed that N-nitrate concentrations behave differently (Fig. 5f). The concentrations in Drogenbos clearly increased between 1990 and 2000 (on average from 2.3 to 3.3 mg L⁻¹), while in Lembeek, the trend is similar but with a much smaller increase. After 2006, the concentrations also increased downstream from Brussels (from 1 to 2.4 mg L⁻¹). This behavior is similar to the one observed for oxygen.

Total phosphorus and phosphate

As for nitrogen, there is a predominant effect of Brussels on the phosphorus content of the river water (on average +1.1 to +2.8 mg L⁻¹ for P_{tot} and +0.3 to +1.5 mg L⁻¹ for P-PO₄, P25% and P75%, respectively, between Drogenbos and Vilvoorde) (Fig. 5g, h).

Upstream from Brussels, total P and P-PO₄ (Fig. 5g, h) decreased in the early 1990s (on average from 1.1 to 0.7 mg L⁻¹ for total P and from 0.7 to 0.3 mg L⁻¹ for P-PO₄), especially in Drogenbos. As already observed for all the other parameters, the decrease downstream from Brussels was most significant between 2006 and 2007 (on average from 3.2 to 1.5 mg L⁻¹ for total P and from 1.3 to 0.5 mg L⁻¹ for P-PO₄).

Discussion

The influence of the river discharge and temperature on the water quality

Considering the long-term trends observed in the Zenne River, variations linked to the hydro-climatic variability (water

discharge and temperature) are certainly not the dominant factors controlling the observed evolution of the BOD, COD, O₂, N, and P concentrations.

Important year-to-year fluctuations of the average annual discharge are however observed in the Zenne River for all stations (Fig. 4), and it might be interesting to explore the influence of these variations on the measured concentrations.

Because of the predominant effect of Brussels on both the water discharge and the concentrations, correlations were considered for the upstream and downstream stations separately. In addition, we also had to take into account changes in the wastewater treatment capacity in the Zenne basin, as this strongly modified the pollutant emissions over the observation period. Indeed, the effects of water discharge variations on the concentration can only be considered for years with similar pollutant (wastewater) loads. Considering the evolution of the wastewater treatment capacity over time (Fig. 2), we selected for each region (up- and downstream from Brussels) three periods of at least 5 years during which the treatment capacity did not significantly change over time. The selected periods for stations upstream from Brussels are as follows: 1990–1999 (treatment capacity 65,000 to 75,000 IE); 2000–2004 (120,000 to 200,000 IE); and 2005–2013 (250,000 to 350,000 IE). For stations downstream from Brussels, the selected periods are 1990–1999 (65,000 to 75,000 IE); 2000–2006 (480,000 to 620,000 IE); and 2007–2013 (1,800,000 to 1,920,000 IE).

Table 1 shows the correlations between selected water quality variables and the discharge for the different periods, up- and downstream from Brussels. For oxygen saturation, significant correlations are observed for all the periods as well as for up- and downstream stations from Brussels. The same is true for N-NH₄ downstream from Brussels, but—if one considers the two more recent periods—not for upstream stations. For total P, P-PO₄, and nitrate, significant correlations may be seen, but not for all the periods and at all the locations. For BOD and total N, no significant correlation was found.

For periods and regions with significant correlation before 2004–2006, average annual oxygen and nitrate concentrations increased with increasing discharge, while average annual COD, N-NH₄, and P-PO₄ concentrations decreased with increasing discharge. Interestingly, a totally different behavior is observed for the most recent period (2005–2013 and 2007–2013) during which average annual oxygen concentrations decreased and ammonium, total P, and phosphate concentrations increased with increasing discharge. This is best illustrated by the oxygen saturation levels downstream from Brussels (Fig. 6).

The most important factors controlling the O₂ levels in rivers are production by photosynthesis performed by phytoplankton, periphyton, and macrophytes; consumption by respiration (mainly bacterial respiration) and nitrification; and exchange with atmosphere (reaeration) (Bennett and

Table 1 Spearman rank order correlations between the average annual water discharge and oxygen saturation ($O_2\text{sat}$), chemical oxygen demand (COD), nitrate ($N\text{-NO}_3$), ammonium ($N\text{-NH}_4$), phosphate ($P\text{-PO}_4$), and total P (Ptot) concentrations

Year	$O_2\text{sat}$	COD	$N\text{-NH}_4$	$P\text{-PO}_4$	$N\text{-NO}_3$	Ptot
Upstream Brussels						
1990–1999	R	0.46	ns	-0.51	ns	ns
	p	0.05		0.02		
	n	18		20		
2000–2004	R	0.71	ns	ns	-0.69	0.79
	p	0.02			0.05	0.005
	n	10			8	10
2005–2013	R	-0.46	0.60	ns	ns	ns
	p	0.06	0.009			
	n	18	18			18
Downstream Brussels						
1990–1999	R	0.50	-0.57	-0.64	ns	0.71
	p	0.03	0.01	0.003		0.0003
	n	18	20	20		20
2000–2006	R	0.76	ns	-0.63	ns	ns
	p	0.002		0.02		
	n	13		13		
2007–2013	R	-0.71	ns	0.68	0.65	ns
	p	0.004		0.01	0.01	
	n	14		14	14	14

Only variables with significant correlations are shown. Upstream Brussels: Lembeek and Drogenbos; Downstream Brussels: Vilvoorde and Eppegem
ns not significant

Rathburn 1972). Before 2004–2006, there was still a massive fraction of untreated wastewater that was discharged to the river (about one third of the present treatment capacity, see Fig. 2) with overall low levels of oxygen as a consequence (Fig. 5c). During this period with very low O_2 levels, the higher oxygen levels observed for higher discharge may be explained by an increase of the reaeration during wet conditions (increased flow velocity and turbulences, rainfall; see the

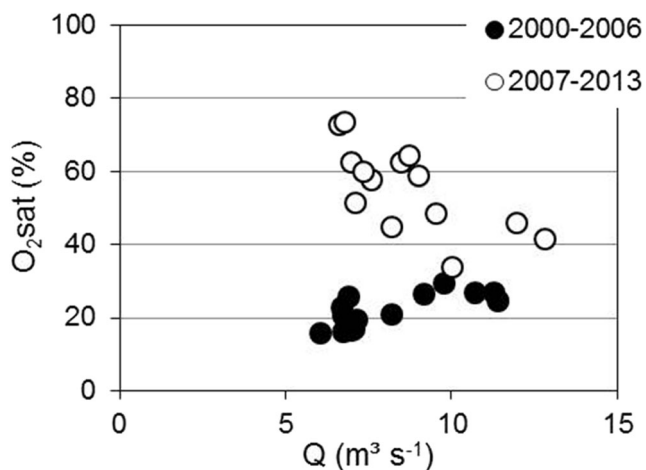


Fig. 6 Average annual oxygen saturation levels ($O_2\text{sat}$) as a function of average annual discharge (Q) downstream from Brussels (Eppegem and Vilvoorde) for the periods 2000–2006 and 2007–2013

review of Cox 2003 on the dependence of the reaeration coefficient towards hydraulic parameters), a more important dilution of the untreated sewage, and a reduced water transit time limiting the significance of O_2 -consuming processes in the river (respiration—pelagic and benthic—and nitrification). However, after 2004–2006, the wastewater treatment capacity was more than doubled, reducing drastically the inputs of untreated sewage, and the river experienced an immediate and strong increase of O_2 levels (Figs. 2 and 5c). In these waters with prevailing better (higher) O_2 conditions, effects linked to reaeration, dilution of (strongly reduced amounts of) untreated wastewaters, and decreased respiration at high discharge do not affect significantly the O_2 levels anymore (O_2 is already high) and other factors may become more important. One of these might be the increased organic matter inputs during wet weather conditions by combined sewer overflows (CSO). Sewer overflows are indeed known to occur very frequently in Brussels (101 events/year for only 1 of the sewer outfalls—Le et al. 2014) and result in decreased O_2 levels downstream Brussels (Le et al. 2014). In addition, for years with prevailing wet weather conditions, the wastewater treatment might be less efficient as the WWTPs are more frequently overloaded and need to use their wet-weather treatment line (primary treatment) more frequently.

Nitrate levels in rivers depend mainly on diffuse sources like groundwater and soil leaching, point sources (mainly

treated sewage from WWTPs with a nitrification stage but no denitrification stage), in-stream production by nitrification, and in-stream consumption by denitrification and plant uptake (Ranalli and Macalady 2010 and references therein). A positive correlation between the NO_3 levels and the river discharge as observed in the Zenne has already been reported in the past (Taleb 1997) and is characteristic for many rivers including rivers from the Scheldt catchment (Struyf et al. 2004). This is explained by the dominance of diffuse sources of nitrate in the catchment—soil leaching and groundwater seepage—that increase during wet weather conditions. These diffuse nitrate sources are most important in the fertilized agricultural areas of the river which are located in the upstream part of the river (see the high nitrate concentrations in Lembeek, Fig. 5f). In addition, as for oxygen, the significance of in-stream retention processes (denitrification and plant uptake) decreases with increasing river discharge because of reduced transit times.

COD, N-NH_4 , and P-PO_4 mainly originate from point sources (sewage) that are diluted during higher flow conditions. In addition, their concentrations can also be impacted by the decreased residence time that is limiting the production of ammonium and phosphate by in-stream mineralization of organic matter. However, as for oxygen, phosphate and ammonium displayed a different behavior after 2007 with an increase of N-NH_4 and P-PO_4 concentrations with increasing discharge. After 2007, the river experienced a strong decrease of P-PO_4 and N-NH_4 levels coinciding with an increased wastewater treatment capacity and tertiary treatment introduction at Brussels North WWTP (Figs. 2 and 5f, h). In these conditions, dilution effects are not visible anymore (river water concentrations are low) and other factors may become more important. For example, the contribution of untreated CSO waters might be important, as suggested before, but a less efficient water treatment by WWTPs during more frequent rain events could play a role as well.

Year-to-year fluctuations of the average annual temperature are much more reduced, and significant correlations with water quality variables are thus only rarely observed. A

significant negative correlation was only observed once with O_2 saturation for the period 2000–2006 downstream from Brussels ($R=-0.55$, $p=0.05$) and illustrates the positive effect of temperature on respiration processes with average lower respiration (higher O_2 saturation) during colder years.

The influence of the wastewater treatment capacity and in stream retention processes on the water quality

At the outlet of the river basin

The obvious influence of the wastewater treatment capacity on the average annual concentrations of the pollutants was considered separately for the up- and downstream stations, and no distinction needed to be made between wet and dry years, illustrating the dominant character of treatment capacity over meteorological controls. The Spearman rank order correlations show that for both the upstream and the downstream stations, the average annual concentrations of all the considered variables were significantly correlated with the wastewater treatment capacity (Table 2). Interestingly, there was also a significant correlation between the treatment capacity and the temperature of the river upstream from Brussels. The correlations were weak ($R<0.5$) for nitrate but strong for all other variables.

The high levels of BOD, COD, total N and P, ammonium, and phosphate in the 1970s and till the early 1990s show the predominant influence of point sources of untreated sewage. Decreasing the sources of untreated sewage by the operation of WWTPs immediately resulted in reduced pollutant concentrations. This evident decrease of organic matter, N, and P concentrations linked to the implementation of WWTP facilities—following the implementation of the urban wastewater Directive 91/271/EEC—was already mentioned in Garnier et al. (2013) and has been observed in many European rivers (Bouraoui and Grizzetti 2011). Directly linked to this, positive correlations were found for both O_2 and nitrate. For oxygen, this shows that the very low yearly average O_2 levels in this

Table 2 Spearman correlation coefficients (R), probability (p), and number of observations (n) between the wastewater treatment capacity and the yearly average temperature (T), oxygen saturation (O_2sat),

biological oxygen demand (BOD), chemical oxygen demand (COD), total N (Ntot), total P (Ptot), ammonium (N-NH_4), nitrate (N-NO_3), and phosphate (P-PO_4) concentration

		T	O_2sat	BOD	COD	Ntot	Ptot	N-NH_4	N-NO_3	P-PO_4
Upstream Brussels	R	-0.64	0.77	-0.51	-0.85	-0.61	-0.54	-0.61	0.36	-0.67
	p	6.0E-7	2.0E-7	5.0E-4	2.0E-7	6.0E-5	7.0E-5	3.0E-6	1.0E-2	1.0E-7
	n	50	48	43	50	38	50	50	50	46
Downstream Brussels	R		0.82	-0.83	-0.82	-0.83	-0.67	-0.72	0.71	-0.84
	p	ns	2.0E-7	2.0E-7	2.0E-7	2.0E-7	2.0E-7	2.0E-7	2.0E-7	2.0E-7
	n		47	44	50	39	49	50	47	47

Upstream Brussels: Lembeek and Drogenbos; Downstream Brussels: Vilvoorde and Eppegem

ns not significant

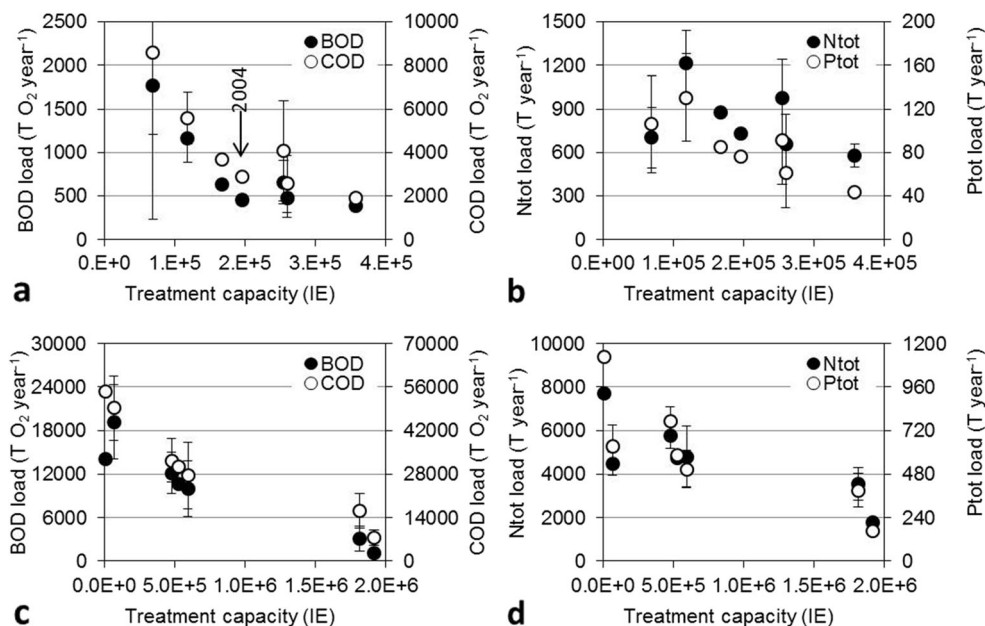
river in the past decades were largely due to the respiration of organic matter from untreated sewage releases. Hence, there is a strong correlation between yearly average O₂ and COD concentrations (Spearman $r=-0.89$, $p=2E-7$) whatever the location considered. Decreasing the sources of untreated sewage by the operation of WWTPs thus immediately resulted in increased oxygen concentrations, demonstrating the fast response of respiration processes towards reduced organic matter inputs as observed in other rivers (for example Daniel et al. 2002). The increased oxygen levels also had an influence on the nitrate dynamics. As discussed before, nitrate in the Zenne basin originates mainly from diffuse sources in the upstream agricultural part of the river basin and, at low levels of oxygen, nitrate is reduced to nitrogen gas by the process of denitrification. The importance of denitrification in the rivers of the upstream basin of the Scheldt for the years 1973–1983 was already demonstrated by Billen et al. (1985). Restoring better oxygen concentrations in this urban river thus decreased its natural capacity of reducing nitrates, with higher NO₃ concentrations as a result. In addition, increased nitrification, resulting from better oxygen conditions, may constitute an additional source of nitrates. Hence, there is an excellent correlation between yearly average N-NO₃ and O₂ concentrations (Spearman $r=0.76$, $p=2E-7$) whatever the location considered. Similar results were found in the estuary of the Scheldt where increased NO₃ fluxes were observed between the 1980s and the 1990s and in the early 2000. These increases were attributed to reduced denitrification/enhanced nitrification caused by better oxygen conditions (Soetaert and Herman 1995; Van Damme et al. 2005). However, upstream from Brussels (Lembek and Drogenbos), the increase of nitrate concentrations due to restored oxygen levels is probably

attenuated by the decrease of diffuse nitrate sources linked to a reduction of N fertilizer application in the river basin. For the Scheldt river basin, it was shown that synthetic fertilizer application decreased from 18,500 kg N km⁻² year⁻¹ in 1990 (Billen et al. 2005) to 5500 kg N km⁻² year⁻¹ in 2000 (Billen et al. 2009), while more local information obtained from the regional administration in charge for agriculture policy in Wallonia DGARNE (Direction Générale opérationnelle Agriculture, Ressources Naturelles et Environnement) shows a smaller decrease of inorganic N fertilizer use from 131 kg ha⁻¹ year⁻¹ in 1999 to 101 kg ha⁻¹ year⁻¹ in 2009 (Marsin, pers. comm.). Hence, annual river N-NO₃ concentrations in Drogenbos (outlet of the upstream Zenne River basin) are relatively stable since 1998 (Fig. 5e) while O₂ saturation levels continued to improve from 40 % (in 1998) to 70 % (Fig. 5c).

Obviously, increasing the wastewater treatment capacity in the Zenne basin decreased the loads of urban pollutants such as BOD, COD, total N, and total P towards the river, and hence decreased the urban pollutant loads transported by the river downstream. This decrease can be quantified at a basin scale by considering the evolution of the annual urban pollutant loads transported by the river at the outlet of the basin as a function of the treatment capacity in the basin. This was done for the upstream part of the basin by considering the annual BOD, COD, total N, and total P loads in Drogenbos, and for the whole basin including the Brussels area by considering the annual loads in Eppegem (Fig. 7).

It is important to note that the observed loads at basin outlets do not only depend on wastewater emissions but are also influenced by other sources of pollutants such as direct industrial wastewater inputs, diffuse inputs such as leaching

Fig. 7 Observed average annual loads for biological oxygen demand (BOD), chemical oxygen demand (COD), total N (Ntot), and total P (Ptot) as a function of wastewater treatment capacity in Drogenbos (outlet upstream basin; **a, b**) and Eppegem (outlet whole basin including the Brussels area; **c, d**). Error bars indicate the standard deviation on the average of years with similar treatment capacity



from agricultural areas, and in-stream retention/production processes such as settlement, respiration, denitrification (for N), and assimilation by aquatic plants (for N and P).

Although there is no quantitative estimation of direct industrial inputs of pollutants in the Zenne basin, these might have been important in the upstream part of the river basin until the 1990s. After this period, the surrounding region experienced a progressive deindustrialization (Mommen 2002) and remaining industrial effluents were all either treated by their own treatment plant or connected to the existing municipal wastewater treatment facilities (and so included in the WWTP treatment capacity, Garnier et al. 2013). The individual contributions of diffuse inputs were not assessed in the framework of this study, but Leta (2013) estimated important diffuse N-NO₃ sources (1200 t N year⁻¹) and more limited diffuse P-PO₄ sources (3 t P year⁻¹) in the upstream catchment of the Zenne basin. Finally, the individual importance of in-stream transformation processes is difficult to assess.

However, the net combined effect of other sources of pollutants and in-stream processes can be estimated by considering the effective specific (per IE) pollutant loads at the outlet of the basin before any treatment (ESP untreated) and comparing this to specific loads known for untreated sewage. Indeed, the difference between both will be the net result of the contribution of all additional pollutant sources (direct industrial inputs and diffuse inputs) and sinks. For this, we considered the local definition of “inhabitant equivalent” from Verbanck et al. (2005) as a daily load per inhabitant corresponding to 54 g O₂ BOD, 120 g O₂ COD, 12 g total N, and 1.45 g total P. It is to be noted that for P, the specific load per inhabitant probably changed over the observation period as it has been influenced by the use of polyphosphates in washing powders in the 1960s (until 4 g P IE⁻¹ day⁻¹) and their progressive removal in the 1990s—the value used here is close to the physiological emission rate of P (1–1.5 g P IE⁻¹ day⁻¹) (Verbanck et al. 1989; Billen et al. 1999; Garnier et al. 2006).

ESP untreated can be estimated by dividing the average annual load observed at the outlet of the basin before any wastewater treatment by the population. As our data do not illustrate a situation without any treatment, we estimated this

ESP untreated by considering the intercept of our data distribution line (Fig. 7) with the Y-axis (loads for IE=0 as treatment capacity) and dividing the intercept with the treatment capacity in 2013. Except for BOD and COD loads in the upstream basin, most of the variables displayed a linear behavior with the treatment capacity over the whole observation period (Fig. 7). For these, ESP was calculated by taking the intercept of a linear regression through all the data. For BOD and COD in the upstream basin, as there is a clear change of slope between the periods 1990–2004 and 2004–2013 (see Fig. 7a), we considered the intercept of a regression for two periods: before 2004 and after 2004. All ESP untreated at the outlet of river basins are lower than standard specific loads of untreated sewage showing a net reduction of pollutant input loads and the dominance of in-stream consumption processes over other sources. Hence, BOD inputs are reduced by 58 to 91 %, COD by 44 to 69 %, total N by 15 to 26 %, and total P by 17 to 21 % (Table 3). For the upstream basin, if we would consider for N and P an extra input by diffusive fluxes of 9 g N IE⁻¹ day⁻¹ and 0.02 g P IE⁻¹ day⁻¹ (calculated from Leta 2013 considering a population of 357,555 IE), the N input reduction would increase up to 51 % while there will be no difference for P. We need to mention that considering the upstream basin, part of the high input reduction may also be attributed to the frequent use of septic tanks by the population before WWTP construction, preventing part of the produced wastewater from reaching the river.

In addition, as demonstrated by the change of slope, the specific BOD and COD loads for untreated conditions strongly decreased between the periods before 2004 and after 2004. This probably shows an important contribution of direct industrial wastewater inputs in the years preceding wastewater treatment (1970s). These direct industrial inputs decreased progressively during the 1990s with the closure of many factories (Mommen 2002), and we observe a much lower ESP untreated for BOD and COD after 2004: respectively 5 and 37 mg O₂IE⁻¹ day⁻¹.

The slope of the data distributions presented in Fig. 7 also informs us about the ESP reduction resulting from increased wastewater treatment capacity, and hence, we can calculate an

Table 3 Effective specific loads (ESP)±SE of biological oxygen demand (BOD), chemical oxygen demand (COD), total nitrogen (Ntot), and total phosphorus (Ptot) at the outlet of the upstream Zenne basin (before and after 2004) and of the whole basin (whole) including the

Brussels area without any wastewater purification. Specific loads in standard untreated wastewater and % difference of ESP compared to standard wastewater

	BOD g O ₂ IE ⁻¹ day ⁻¹	COD g O ₂ IE ⁻¹ day ⁻¹	Ntot g IE ⁻¹ day ⁻¹	Ptot g IE ⁻¹ day ⁻¹
Upstream basin before 2004 (1)	19±1	87±6	10±1.6	1.2±0.2
Upstream basin after 2004 (2)	5±2	37±17	10±1.6	1.2±0.2
Whole basin (3)	23±2	67±5	8.8±0.9	1.2±0.1
Untreated wastewater	54	120	12	1.45
% difference for (1), (2), and (3)	(-65), (-91), and (-58)	(-27), (-69), and (-44)	(-15), (-15), and (-26)	(-21), (-21), and (-17)

effective “efficiency” of BOD, COD, Ntot, and Ptot removal induced by wastewater purification at a basin scale. It is to be noted that this efficiency also includes other changes such as reduced direct industrial inputs or increased in-stream retention process. This effective treatment efficiency (ETE in %) can be calculated by comparing the observed ESP reduction (slope) to the ESP in the situations with no wastewater treatment (Table 3). For BOD and COD in the basin upstream from Brussels, as noticed before, two periods with different slopes need to be distinguished (Fig. 7a) (before 2004 and after 2004), while for other distributions, the whole observation period was considered.

Observed ESP reductions in the whole basin (Table 4) compared to ESPs in the situations with no wastewater treatment showed ETEs of 90 % for BOD, 80 % for COD, 59 % for total N, and 73 % for total P. These are very close or even higher than what is usually required for WWTPs in Europe (at least 70 % for BOD, 75 % for COD, 70 % for total N, and 80 % for total P—according to the lowest requirements of the Urban Wastewater Directive 91/271/EEC). Similarly, the ETEs of nitrogen (73 %) and phosphorus (53 %) loads in the upstream basin are very close to expected WWTP efficiencies. It is to be noted that for P, part of the observed ETE might also be due to the obligate removal of polyphosphates from detergents since the early 1990s (see also discussion in “In the Brussels area”).

The observations are however quite different for BOD and COD in the upstream basin before and after 2004. Before 2004, in the upstream basin, the ETEs calculated for BOD and COD are higher than 100 %. These very high BOD and COD load reductions before 2004 can mainly be attributed to the added contribution of the progressive deindustrialization of the region of the upstream basin of the Zenne during the 1990s with the closure of many factories (Mommen 2002), and hence reduced industrial loads of organic matter. We indeed observed a strong reduction of ESP untreated for BOD and COD between the periods before and after 2004 (Table 4): 14 g O₂IE⁻¹ day⁻¹ for BOD and 47 g O₂IE⁻¹ day⁻¹. After 2004, increasing the WWTP capacity from 2 × 10⁵ to 4 × 10⁵ IE upstream from Brussels did not result in important COD and BOD load reductions anymore (ETEs are not significantly different from zero) (Fig. 7a and Table 4). Most

probably, after 2004, the lower remaining BOD and COD loads associated to untreated sewage emissions were reduced to their lowest levels by the high self-cleansing in-stream processes (more than 90 % for BOD and 69 % for COD, see Table 3) before reaching the observation station of Drogenbos. Increasing treatment capacity did thus not result in visible BOD and COD load reduction anymore but contributed to reduced in-stream respiration processes as illustrated by the increased O₂ saturation observed after 2004 while BOD and COD remain low (Fig. 5a–c). Remaining loads probably correspond to the less biodegradable organic matter fraction that is only slowly degraded. Indeed, from the early 1990s until 2004, the observed ratio of BOD/COD which can be used as an indicator of the biodegradability of organic matter (Vollertsen and Hvitved-Jacobsen 2002) decreased from 0.28 to 0.18 and is stable until now (between 0.15 and 0.21 for the period 2004–2013).

In the Brussels area

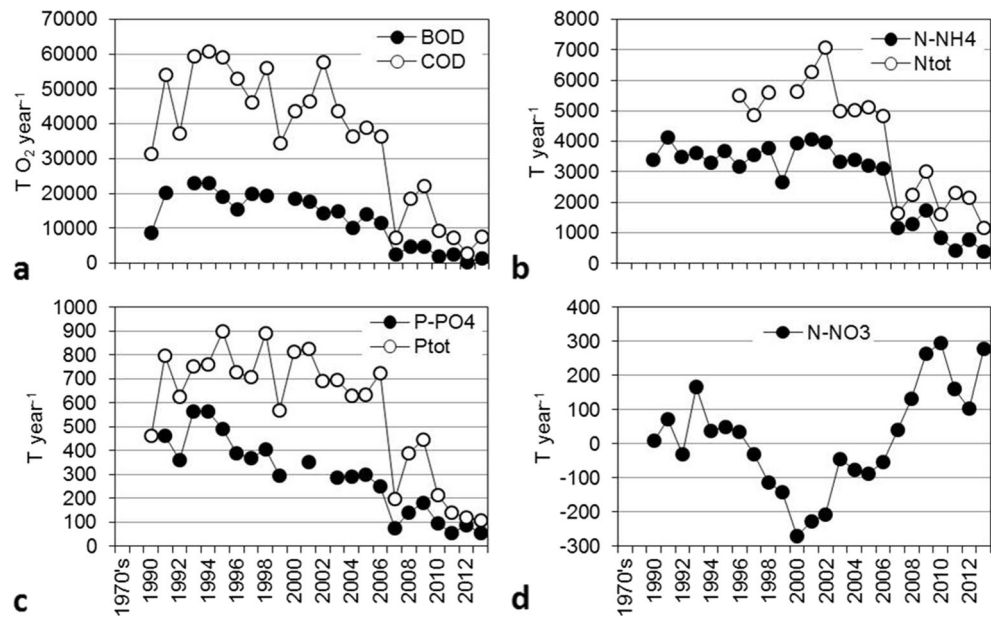
As the population in the basin mainly concentrates in the vicinity of Brussels, the effects of wastewater management actions in and around the city are the most striking, as illustrated by the different levels of concentrations in the upstream and the downstream stations. The differences between the loads at stations Drogenbos and Vilvoorde allow a direct estimation of the quantitative input of organic matter and nutrients from the city to the river and its evolution over time (Fig. 8). Indeed, most of the main sewers of the city and the present-day WWTPs discharge within the river stretch that is delimited by these two stations except for a small fraction (ca 130,000 IE until 1999 = 6 % of total Brussels wastewaters). The inputs of COD, BOD, total N, total P, and ammonium were very high (loads are multiplied by 10 for COD, total N, total P, and N-NH₄, and by 20 for BOD) and relatively stable until 2006, then decreased abruptly in 2007 when the largest Brussels North WWTP started to be operational. It is to be mentioned that although about 99 % of the wastewater is now treated in Brussels, the loads in Vilvoorde are still about five (for COD, total N, total P and N-NH₄) to ten (for BOD) times higher than in Drogenbos. The inputs of phosphate by Brussels were also high (loads multiplied by 10 until 2006)

Table 4 Effective treatment efficiency (ETE)±SE of biological oxygen demand (BOD), chemical oxygen demand (COD), total nitrogen (Ntot), and total phosphorus (Ptot) at the outlet of the upstream Zenne basin and of the whole basin including the Brussels area

	BOD %	COD %	Ntot %	Ptot %
Upstream basin (before 2004)	-154±13	-141±18	-53±22	-73±23
Upstream basin (after 2004)	Ns0	Ns0	-53±22	-73±23
Whole basin	-90±15	-80±15	-59±20	-70±23

Ns0 not significantly different from 0

Fig. 8 Difference between the average annual loads in Vilvoorde and Drogenbos of **a** biological oxygen demand (*BOD*) and chemical oxygen demand (*COD*), **b** total nitrogen (*N_{tot}*) and ammonium (*N-NH₄*), **c** P-phosphate (*P-PO₄*) and total phosphorus (*P_{tot}*), and **d** nitrate (*N-NO₃*), as a contribution of the Brussels area



but started to decrease in the early 1990s. As there was no sewage water treatment in Brussels at that period, this early decrease can be attributed to the obligate removal of polyphosphates in detergents during these years (for the Scheldt, see for example Zwolsman 1994). Unpublished results by the authors show that median daily P-phosphate loads by Brussels untreated sewage indeed decreased from 1020 kg day^{-1} in 1992 to 730 kg day^{-1} in 2008–2010. Presently, the P-PO_4 river loads are still multiplied by 5 when crossing Brussels, showing that there are still important sources of P in domestic sewage. Recently, Comber et al. (2013) reevaluated the modern main contributors to the domestic P load in UK: natural human diet (40 %), food additives (29 %), automatic dishwashing detergents (9 %) and domestic laundry (14 %), and other minor contributors (8 %).

The calculated loads between Drogenbos and Vilvoorde do not only include wastewater inputs (treated or untreated) but also two tributary river contributors: the Zuunbeek and Woluwe rivers (see map in Fig. 3). Zuunbeek and Woluwe rivers are both located in urban areas (northeastern Brussels for the Woluwe and southwestern Brussels and suburbs for the Zuunbeek) and were used as sewers until the recent operation of Brussels North WWTP (in 2007) for Woluwe and St Pieters-Leeuw (in 2005) for Zuunbeek. Their own contribution to the pollutant load is thus only relevant after 2007 for Woluwe and after 2005 for Zuunbeek. For these periods, we calculated that the contribution of the Zuunbeek represented on average 15, 6, 8, 10, and 10 % of respectively the water, BOD, COD, N_{tot} , and P_{tot} load increase observed in the Brussels area, while the Woluwe contribution was of 3 % for the water discharge and 2 % for whatever pollutant considered. We can thus say that tributaries are not important as pollutant

sources for the river between Drogenbos and Vilvoorde compared to wastewaters.

In addition, there are also the contributions of in-stream transformation processes such as organic matter mineralization and settlement of particulate material. As seen before, in-stream transformation processes do occur and might significantly affect concentrations (and thus loads). However, as treated wastewaters represent on average half of the water downstream from Brussels, one can reasonably assume that their effect is dominant. Evidence for this can be found when dividing the obtained loads by the Brussels population for the corresponding years (population in the Brussels region per year from 964,000 in 1990 to 1,155,000 in 2013, Statistics Belgium). Before 2007, the calculated daily loads per inhabitant were on average 40 g of BOD, 122 g of COD, 16 g of total N, 10 g of N-NH_4 , 2 g of total P, and 0.9 g of P-PO_4 . After 2007, the daily loads per inhabitant were on average 6.6, 27, 5.1, 0.6, 2.4, and 0.2 g for BOD, COD, total N, total P, N-NH_4 , and P-PO_4 , respectively. These values of per capita loads are within the ranges usually found for raw (before 2007 period) and tertiary treated sewage (after 2007 period) in comparable combined sewer systems (Servais et al. 1999).

The behavior of nitrate is totally different (Fig. 8). Nitrate content is known to be low in O_2 -depleted untreated sewage. Before 1996 (no wastewater treatment in Brussels), nitrate is low in the river upstream (between 1 and 2 mg N L^{-1}) because of reduced O_2 levels, and the input of nitrate by Brussels is very low and almost zero. From 1997 till 2007, there is a small sink of nitrate (about 10 % of the incoming nitrate loads) in the 20-km Brussels river stretch. In this period, nitrate concentrations increase upstream from Brussels due to better O_2 levels (Fig. 5e) while Brussels sewage is still untreated (or partly treated after 2000) and O_2 levels in the river water in

Brussels are very low (yearly average around 2 mg L⁻¹, see Fig. 5c), which allows denitrification to occur (see previous part of the discussion). It is only after 2007 that the input of nitrate becomes more important and loads are then multiplied by 1.6. The small input of nitrate may be attributed to the fact that part (one fourth) of the wastewater of Brussels only undergoes secondary treatment (so no N retention).

Present status of the water quality of the Zenne River according to European criteria

Nowadays, most of the wastewater produced in the area of the Zenne basin is treated before its disposal to the river, and we have shown that Zenne River water quality experienced a clear amelioration as a response to domestic wastewater treatment. However, one of the remaining questions is whether this better water quality is now sufficient to consider the river is in a good status. According to the European Water Framework Directive (2000/60/EC), all European rivers (and other water bodies) should have a “good status” or, in the case of strongly modified water bodies such as the Zenne River, a “good potential” by 2015. This status is defined by comparing the present biological, chemical, and morphological conditions with the ones prevailing in the system for a reference situation with minimum human disturbance. The definition of this reference situation is type/regional specific and has lead national—or regional in the case of Belgian environmental policy rules—authorities to define a series of biological, chemical, and morphological criteria values or norms to respect. Regarding the variables considered in this paper, and the location of our monitoring stations (in Flanders region, see Fig. 3), the criteria and their target values to respect are defined by Flanders authorities for rivers of the type “large rivers.”

For this type of rivers, and regarding our variables, good water quality needs to respond to the criteria values defined in

Table 5: for oxygen, the 10th percentile of the yearly distribution needs to be higher than 6 mg L⁻¹; for BOD, COD, and N-NO₃, the 90th percentile of the yearly distribution needs to be lower than 6, 30 mg O₂L⁻¹, and 10 mg N L⁻¹, respectively; for ammonium, the median of the yearly distribution should be lower than 2 mg L⁻¹; for phosphate, the average of the yearly distribution should be lower than 0.1 mg L⁻¹; and for total N and P, the average of “summer” months (April, May, June, July, August, and September) should be lower than 4 and 0.14 mg L⁻¹, respectively. All criteria values were calculated for the combined years 2011–2013 at our four monitoring stations (Table 5). According to this definition, none of the considered stations in the Zenne River are fulfilling the requirements for oxygen, total nitrogen, total phosphorus, and phosphate concentrations. Ammonium, COD, and BOD also often exceed the target value except in Lembeek, but their excess is more limited when compared to the O₂, N, and P criteria. Only nitrate levels are satisfactory at all stations.

The Zenne waters before Brussels (Drogenbos) are already largely contaminated by N, and P, and in a lesser way by BOD and COD. Part of this contamination might be explained by the high population density and high contribution of treated wastewaters to the river flow. We estimated the potential contribution of treated wastewater sources on the concentrations measured at Drogenbos by considering a wastewater volume input of 120 L IE⁻¹ day⁻¹, as defined by the Walloon and Flanders region authorities, and an average treated wastewater composition for the effluents of WWTPs. This mean composition was calculated from real observations as the weighted average (Table 6) of the effluent characteristics of each WWTP located in the upstream basin (data obtained from the wastewater treatment agencies SPGE and AQUAFIN for the Walloon and Flanders regions, respectively). WWTP effluents contribute on average to 16 % of the river discharge in Drogenbos (Table 6), and the amounts of BOD, COD, Ntot,

Table 5 Criteria and target value defining good water quality for oxygen (O₂), biological oxygen demand (BOD), chemical oxygen demand (COD), ammonium (N-NH₄), nitrate (N-NO₃), total nitrogen

(Ntot), total phosphorus (Ptot), and phosphate (P-PO₄) and their values for the combined years 2011–2013 in Lembeek, Drogenbos, Vilvoorde, and Eppegem

Variable	Criteria	Target	Lembeek (2011–2013)	Drogenbos (2011–2013)	Vilvoorde (2011–2013)	Eppegem (2011–2013)
O ₂ (mg L ⁻¹)	P10%	>6	4.5	5.1	5.6	3.1
BOD (mg O ₂ L ⁻¹)	P90%	<6	4.6	7.0	7.0	5.0
COD (mg O ₂ L ⁻¹)	P90%	<30	28	31	39	37
N-NH ₄ (mg L ⁻¹)	Median	<2	1.0	1.3	2.8	3.1
N-NO ₃ (mg L ⁻¹)	P90%	<10	4.9	5.1	3.4	3.9
Ntot (mg L ⁻¹)	Summer ^a average	<4	5.6	7.0	9.9	8.0
Ptot (mg L ⁻¹)	Summer ^a average	<0.14	0.5	0.6	0.8	0.8
P-PO ₄ (mg L ⁻¹)	Average	<0.1	0.2	0.6	0.4	0.4

Italicized values are satisfying the target criteria value

^a Summer=months April, May, June, July, August, and September

Table 6 Average discharge (Q), biological oxygen demand (BOD), chemical oxygen demand (COD), total nitrogen (N_{tot}), total phosphorus (P_{tot}), ammonium (N-NH₄), and phosphate (P-PO₄) concentrations in treated domestic wastewater (WW) and in Zenne River waters at Drogenbos; total amount of water and pollutants added by WWTP effluents to river waters

	Average treated WW	Drogenbos (2011–2013)	Added by WWTP
Q (m ³ s ⁻¹)	0.5	2.8	0.50
BOD (mg O ₂ L ⁻¹)	4.9	4.6	0.87
COD (mg O ₂ L ⁻¹)	35	23	6.3
N _{tot} (mg L ⁻¹)	9.7	7.1	1.7
P _{tot} (mg L ⁻¹)	0.85	0.6	0.15
N-NH ₄ (mg L ⁻¹)	1.0	1.5	0.19
P-PO ₄ (mg L ⁻¹)	0.84	0.6	0.15

P_{tot}, ammonium, nitrate, and phosphate added to the river are relatively small compared to observed concentrations in Drogenbos (four to eight times lower, Table 6). Treated wastewaters do contribute to the excess pollution levels observed in Drogenbos, but they are not the dominant sources. Other sources such as diffuse inputs by, for example, soil leaching are presently probably more important in the upstream basin of the river which has a substantial area of agricultural lands, especially for N and P.

Downstream from Brussels, the situation is becoming more critical and we know that treated wastewater from Brussels is a substantial part (50 % on average) of the Zenne River discharge in Eppegem. As done for Drogenbos, we can quantify the specific contribution of WWTP effluents as pollutant sources for Zenne River waters by considering the average treated wastewater volumes and composition of all WWTPs located between Drogenbos and Eppegem (obtained from the wastewater treatment agencies SBGE and AQUAFIN for the Brussels and Flanders regions, respectively). Results (Table 7) show that presently downstream Brussels, treated wastewaters are clearly the dominant source for all considered pollutants with added concentration values similar to the concentrations

Table 7 Average discharge (Q), biological oxygen demand (BOD), chemical oxygen demand (COD), total nitrogen (N_{tot}), total phosphorus (P_{tot}), ammonium (N-NH₄), and phosphate (P-PO₄) concentrations in Zenne River waters at Eppegem; total amount of

Average	Eppegem (2011–2013)	Added from upstream	Added by tributaries	Added by WWTP
Q (m ³ s ⁻¹)	7.6	2.8	0.75	4.4
BOD (mg O ₂ L ⁻¹)	4.8	1.7	0.73	5.7
COD (mg O ₂ L ⁻¹)	34	8.3	3.0	32
N _{tot} (mg L ⁻¹)	8.6	2.6	0.87	5.6
P _{tot} (mg L ⁻¹)	1.0	0.20	0.09	0.68
N-NH ₄ (mg L ⁻¹)	2.9	0.54	0.41	2.2
P-PO ₄ (mg L ⁻¹)	0.4	0.20	0.05	0.36

observed in Eppegem. Compared to the contribution of upstream waters and tributaries (Zuunbeek and Woluwe), WWTP effluents are two to four times more important. In addition, we should also mention that the bad oxygen levels downstream Brussels might also be influenced by the high frequency of CSO events occurring in Brussels. Indeed, as mentioned before, a recent study made by Le et al. (2014) showed for one of the main sewer outfalls in Brussels a CSO frequency of 101 events per year with 73 of these events resulting in oxygen depletion in Eppegem.

Conclusions

The analyses of a 40-year data record of organic matter and nutrients in a highly urbanized stream allow drawing some conclusions that may apply to other highly urbanized river systems. In this type of systems, the average annual river water quality is dominated by urban wastewater disposal and by the progressive installation of wastewater treatment plant facilities, themselves being strongly dependent on regional environmental policy decisions. The presence of a large city on the course of the river has a predominant importance as a source of organic matter and reduced forms of N and P, as shown by the influence of Brussels. Self-purification of domestic loads within the river basin can be very important especially for organic matter loads (important respiration) and may lead to reaching minimum levels of BOD and COD in river waters before having a 100 % treatment capacity, as shown for the upstream Zenne River basin. In addition, they control the oxygen levels largely depending on the respiration of organic matter and influence the nitrate levels. Nitrates mainly originate from diffuse sources and hence depend on fertilizer application in the agricultural areas of the basin, but their distribution in the river downstream is largely correlated with the oxygen level which controls the processes of denitrification and nitrification. Observed decreases of annual pollutant loads at the outlet of the river basin are relatively comparable to what is expected from the implementation of

water and pollutants added by WWTP effluents and tributaries (Zuunbeek and Woluwe) to river waters; and amount of water and pollutants coming from upstream

WWTPs in the basin. In correlation, oxygen levels and nitrate loads increased accordingly. Rainfall conditions influencing average annual river discharge have an influence on water quality, especially on the oxygen levels. If the river is dominated by the release of untreated sewage, as it was the case in the Zenne before the mid-2000, the influence is positive probably mainly because of the dilution of point source pollutant loads. When wastewaters are treated, the influence is negative probably because of less efficient wastewater treatment during wet weather conditions and the more frequent occurrence of combined sewer overflows. Finally, we also conclude that even with the implementation of wastewater treatment, it is very difficult to reach a good status in highly urbanized streams, because of important diffuse sources in agricultural areas, such as in the Zenne basin upstream Brussels, or because of the high volumetric proportion of treated wastewaters—which in spite of their treatment, do not have the quality standards of river waters—as observed at the outlet of the whole Zenne River basin under the dominant effect of the Brussels area. In addition, the frequent occurrence of CSOs in large cities might also contribute to the low O₂ levels observed at the outlet of the basin. In these conditions, it appears that detailed investigations are needed to explore additional actions that could improve water quality in urban streams. Better water quality could be obtained by adopting existing improved wastewater treatment technologies (based on membrane techniques, for example) and by decreasing CSO frequency (improved sewer systems, increased volumes of storm water storage basins, decreasing surface runoff by promoting direct infiltration techniques). But these technological solutions are generally expensive and probably financially hard to support by public budgets. Hydrological solutions such as the reinforcement of water flows during critical summer low-flow conditions, additional land use regulations to mitigate diffuse pollution sources, and restoration of natural/controlled wetlands with a high self-purification potential could be other interesting measures that need to be investigated if we want to reach the good status in urban river waters.

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