

Impact of the hydrological behaviour on pollutant dynamics in an intermittent Mediterranean river (Vène, France).

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ABSTRACT

Intermittent Mediterranean rivers show long draught periods interrupted by floods of high intensity and short duration. Their channel network structure is also highly variable in space and time. Water quality dynamics in these rivers is highly impacted by this specific hydrological behaviour.

During the dry period, the river channel may dry up completely except in some reaches where anthropogenic point-source inputs (mainly waste water treatment plant effluents) contribute to maintain pools. Accumulation of pollutants is observed in these reaches. During the first floods, flow is rarely continuous all along the channel network, because of the existence of transmission losses. At the beginning of a storm, propagation of flood waves is limited by infiltration in the channel bed, but also by filling-up of localized storage zones. Total flood volumes therefore do not increase very much, and even decrease downstream. Some floods observed in the upstream part of the river are not transferred to the outlet. This, in turn, influences the spatial and temporal dynamics of pollutant fluxes. Along the river course, pollutograph shapes show significant differences. Intense autumn floods flush away suddenly the pollutants from the soils and the river bed, but large amounts of pollutants remain trapped in the riverbed, if transmission losses are significant.

Due to this peculiar Mediterranean hydrological regime, specific observation networks must be set up to assess the temporal and spatial variability of pollutant fluxes in the river, the processes governing pollutant transport, as well as the ranking of the origins of the pollutants.

All these phenomena are illustrated here on the case of the Vène River (67 km², South of France) over the observation period: from September 2003 to August 2006, on suspended solids, nutrients and faecal bacteria. The combination of continuous and event based monitoring coupled with the multi-scale sampling strategy allows a global understanding of the factors and processes influencing water quality at the catchment scale: concentration data are used to assess the contamination levels of the river and the bio-availability of pollutants, while by analysing the instantaneous fluxes the various contributions to the river might be established. Finally total flood loads are calculated in order to evaluate the exports at the catchment outlet. The multi-disciplinary approach followed herein improved our knowledge of the integrated catchment response and its impact for downstream environments.

KEYWORDS Water quality, pollutant loads, dry period, first flush flood effect, accumulation, perceptual model

INTRODUCTION

The dynamics of chemical or bacterial contaminants of a river depend on whether inflows of elements into the river are direct or diffuse, their transformation in the river and their transport along the river's course (Haycock et al., 1993; Fischer et al., 1998)

With the exception of certain catchments with particular lithologies or and with the exception of particular elements (e.g. iron), man is most often responsible for the pollutants found in rivers. This is particularly true of the contaminants studied here: nutrients, the majority of which result from fertilization, animal rearing, certain industrial activities, and so-called domestic wastes, as well as bacteria that provide evidence of faecal contamination of human or animal origin. Point inflows of waste water treatment plants are the main source of phosphorus pollution, particularly organic phosphorus, but also of considerable quantities of faecal bacteria. These inflows are usually continuous and only vary at a very small time scale (hourly or daily). In contrast, diffuse inflows are linked to flushing or leaching of soils: the pollutants are transferred to the drainage network, mainly during rainfall events (Sharpley & Syers, 1979). Then, hydroclimatic conditions, in particular the intensity of rainfall events, and the hydrological response of the catchment, thus drive the transfer of the pollutants to the river.

The velocity of transfer of pollutants from their point of emission to the outlet is controlled not only by the hydrological response of the catchment to rainfall events, the ways runoff follows to the river, its propagation in the drainage network, but also the length of the preceding dry period. The hydrological and hydraulics conditions play a major role. During rainfall events, runoff and floods facilitate the evacuation of pollutants downstream and cause rapid variations in concentrations and flows at time scales of often less than one hour (Meybeck, 2005). But in periods with no rainfall, the reduction in flow results in the immobilization of the pollutants in the catchment and in the river bed, and increases biological-physical-chemical processes, which are determined by environmental conditions (Baldwin & Mitchell, 2000). So, on their way from their sources to the drainage network and finally to the outlet, the pollutants undergo successive cycles of accumulation and transport. These cycles have been very well described in the case of faecal bacteria by Crowther et al. (2002). For nutrients, even if most studies were concerned with sites in temperate climates, a few authors have focussed on this subject, particularly in Spain, for example Avila et al. (2002), Ramos & Martinez-Casasnovas (2004), Bernal et al. (2005).

The length of the dry period determines the quantities of pollutants that are likely to be stored in the soil or in the river bed. Even though this is not specific to Mediterranean catchments, it is particularly significant in a region where drought conditions can last several months and where direct inflows represents the major part of the runoff during low flow conditions. The very low speeds involved do not enable mobilization, or even dilution of the pollutants; on the contrary, they facilitate immobilization (Jamieson et al., 2004) and many bio-chemical reactions then occur that modify the bioavailability of the elements (Sigg et al., 2006). The trapping of nutrients in sediments and in biofilms during low flow periods was demonstrated almost 20 years ago (Dorioz et al., 1989; Triska et al., 1989), although trapping is reduced immediately downstream from the direct inflows, due to overloading (Marti et al., 2004). Nitrification and denitrification processes reduce the amount of nitrogen available in the water column (Brezonik, 1997). Some nutrients are assimilated by the aquatic biomass (Marti & Sabater, 1996). Biofilms are formed, and this process is facilitated by higher temperatures and the increase of solar radiation in summer. Stocks of faecal bacteria accumulate, and can survive long periods (Crabill et al., 1999) thanks to high rates of organic matter. But when the water courses and the sediments dry out, the chemical, organic and mineral balance is modified (De Groot and Van Wijck, 1993; Baldwin & Mitchell, 2000; Golterman, 2004), as is the bacterial biomass, which may multiply, as in the case of faecal bacteria (Sherer et al., 1992).

The first rainfall events and the associated floods are considered to be a critical period in the hydrological and hydrochemical functioning of intermittent rivers (Durand et al., 1993). The rain immediately mobilizes the pollutants that have been accumulated on soils in the watershed and transfers them to the drainage network. In this way erosion of agricultural land induced by surface runoff removes the elements adsorbed on soil particles, which is particularly true in the case of faecal bacteria (Tyrell and Quinton, 2006), organic nitrogen and phosphorus, even a long time after the fertilizer has been spread, as shown in the Mediterranean region by Ramon & Martinez-Casasnovas (2006). Moreover the rain that washes urban surfaces can supply a large proportion of nutrients even in catchments essentially devoted to agriculture (Dorioz et al., 1989) and in Mediterranean areas (Asaf et al., 2004). In the case of repeated rainfall events, concentrations decrease rapidly due to exhaustion

of the stocks, as demonstrated by many authors, in particular in the Mediterranean region (Massoud et al., 2006).

As the river network fills with water, the elements that accumulated in the sediments and/or biofilms are also picked up and transported by the flood wave. The contribution of the “riverbed-reservoir” to the exported mass during the first floods has been widely demonstrated (Crabill et al., 1999; House & Denison, 2002; Muirhead et al., 2004). Of course, the transport of mobilized elements also depends on water conditions, i.e. on the degree of flooding and the continuity of runoff, but also on physico-chemical conditions and on the bioavailability of the elements.

This paper aims to explain the dynamics of nutrient and faecal bacteria in the Vène River, during the low flow period and the first autumn flood. The analysis is based on observed data collected during the 2003: (1) in steady state conditions, during the low flow period; (2) in unsteady state conditions, during the first autumn flood in 2003. Concentration and load patterns in steady state conditions are compared. The rates of accumulation of the pollutants in the riverbed are estimated, as well as the flush effect of the first autumn flood.

MATERIALS AND METHODS

The study was conducted on the Vène basin, located in South France. The location, the main land use characteristics and the observation network are presented on Figure 1

Site description

The river Vène drains a superficial basin of 67 km² that ranges from 2 to 323 m in elevation. It is fed by two springs flowing out a Jurassic karst: Cournonsec spring upstream and Issanka spring in the lower part of the river. The river has a 12-km course with a regular slope of 0.4% and a Strahler stream order of 3.

The Vène catchment is sparsely populated; about 12,700 inhabitants are living in three small villages, which cover 3% of the total basin area. In terms of land use, 63% of the total area corresponds to natural karstic zones, covered by garrigue and pine trees. The central part of the basin is flat and devoted to agricultural activities, vineyards represents over 21% of the total basin area.

Point-source pollutant inputs mainly come from three sewage treatment plants (STPs) that are sized for 6450 equivalent-inhabitants. Their effluents have a strong seasonal variability, due to the extensive treatment process.

Observation networks

Four rain gauges are installed in the catchment area. Rainfall data are collected from tipping-bucket devices and recorded continuously during rainfall events.

Three stream gauges are installed along the river: the station *V* is located at the outlet of the basin; the station *K* gauges the high karstic spring plus a small rural catchment (area 1.4 km²); the station *S* defines a nested catchment between the spring and the outlet and drains a catchment area of 35 km². Water level data are recorded on a 5-minute time step. Rating curves are available for the three stations.

A refined observation network is designed for one-day survey campaigns. Ten stations are located along the main river. They include the three stream gauging stations: *V*, *S* and *K*, plus seven sites located upstream and downstream the main natural or artificial tributaries: they are numbered downward *R1* to *R7*. Eleven stations control the main point-source inputs or tributaries. They are numbered downward *T1* to *T11*: *T1*, *T5*, *T10* correspond to sewage treatment plants; *T2*, *T3*, *T4*, *T6*, *T7*, *T8*, *T9* are the main tributaries; *T11* control the lower karstic spring.

Measurement and sampling protocols

Two types of campaigns have been set: one-day survey campaigns that are conducted in steady flow conditions and event campaigns that are conducted during floods.

During the one-day campaigns, the same protocol was followed at each station of the refined network. Water discharge was gauged by velocity field exploration, using a currentmeter; the results are given with $\pm 5\%$ error in these specific flow conditions. Physical and chemical parameters (water conductivity, dissolved oxygen, redox potential, temperature and pH) were measured *in situ* with specific probes. Water samples were taken manually from the middle of the flow section, kept in cool box and carried back to the laboratory within less than four hours. The samples were immediately treated and preserved until the analysis.

During event campaigns, water samples were automatically collected at the three gauging stations V, S and K, on an hourly basis, with an automatic water sampler coupled to the stream gauges. Water conductivity and temperature were measured *in situ* with specific probes and recorded using the same time step. Every six hours, the samples were taken out of the samplers and carried back in cool boxes. At the laboratory, the samples were immediately treated and preserved until the analysis.

Routine samples are collected at the three gauging stations, twice a month.

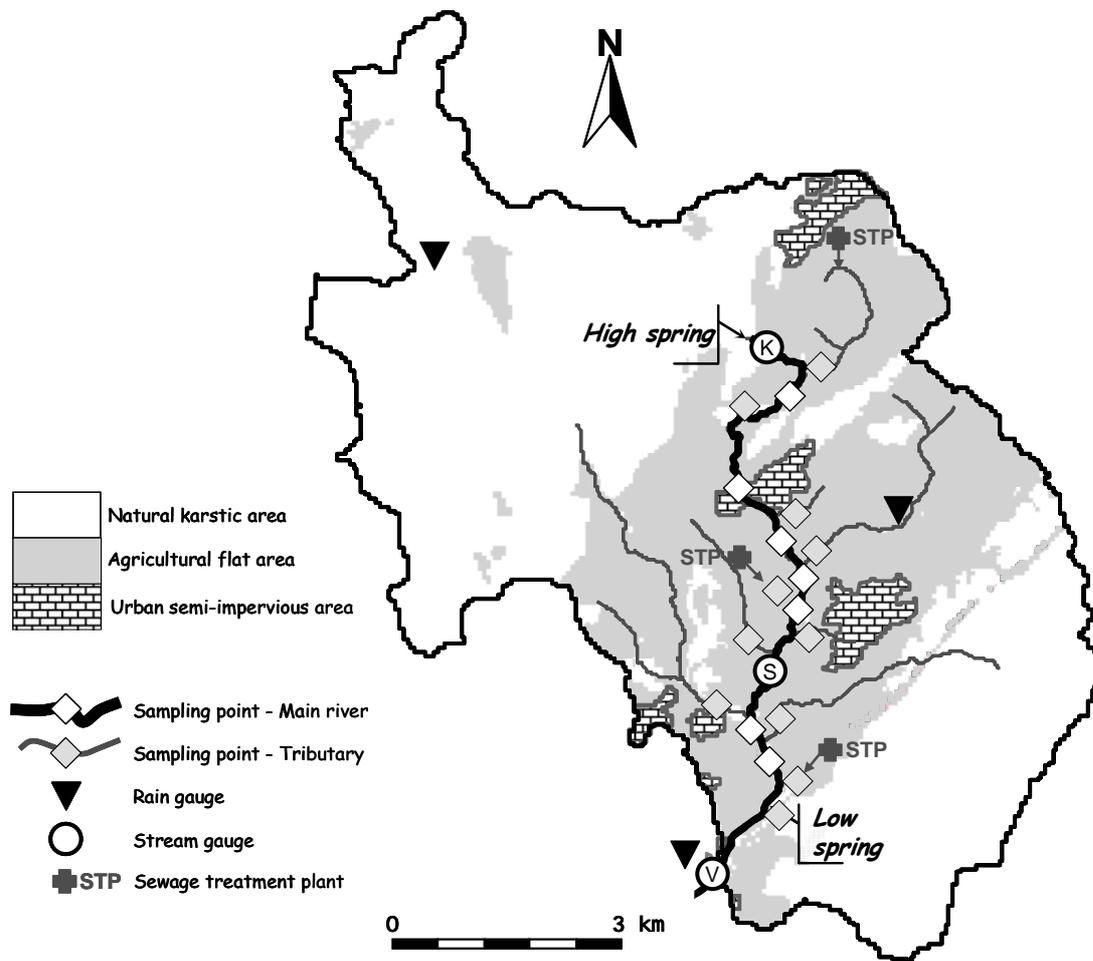


Figure 1 Vene river - land uses and observation network

Nutrient and faecal bacteria determinations

Water samples were analysed for total suspended solids (TSS), ash free dry mass (AFDM), total phosphorus (TP), soluble reactive phosphorus (SRP), ammonium ($\text{NH}_4\text{-N}$), nitrate+nitrite ($\text{NO}_x\text{-N}$), Kjeldahl nitrogen, thermotolerant coliform (TTC) and faecal streptococci (FS).

All the analyses were done following the Standard Methods requirements (APHA, 1992).

Total suspended solids were determined on Whatman GFF filters; 50 mL to 2 L of water were filtered depending on the solid load. Ash free dry masses were obtained after igniting the filters at 550 °C during 1 hour. Concentrations are expressed in mg/L.

Soluble reactive phosphorus was analysed from filtered samples using molybdate blue method. For the determination of total phosphorus, the unfiltered samples were digested by potassium peroxodisulphate before analysis with ammonium molybdate. Concentrations of phosphorus are expressed in µgP/L.

Ammonium was analysed using the spectrophotometric indophenol blue method. NO_x-N was analysed on filtered samples by reduction of nitrates in Cu-Cd column, followed by nitrites determination. Nitrite was determined after normalization (ISO 13395). Total nitrogen was defined as the sum of Kjeldahl nitrogen and NO_x-N. Concentrations of nitrogen are expressed in µgN/L.

Abundances of faecal bacteria were determined by standard membrane filtration method, with two replicates. The results are expressed in colony forming units per 100mL (cfu/100mL).

Load calculations

For the one-day survey campaigns, suspended solids and nutrient loads carried by the main river or brought by the point-source inputs are calculated just by multiplying concentration and discharge values at each point. The results are used to evaluate the mass-balance within the river reaches. For a given reach, the mass-balance is defined, as done by Bowes and House (2001), as the stock of pollutant within the reach i.e the sum of upstream pollutant load, plus loads brought by the point-source inputs, minus downstream pollutant load. Accumulation, retention or fate of pollutants is likely to occur within a reach with a positive mass-balance.

For the event campaigns, total flood loads are evaluated using an integration process on the continuous discharge values (Salles *et al.*, 2008), assuming that concentrations are constant between two samples.

RESULTS AND DISCUSSION

The main features of the dynamics of nutrients and faecal bacteria in the Vène rive are assessed between April and October 2003. Three one-day survey campaigns were conducted in June, September and October 2003. The first autumn flood was sampled at the three gauging station.

General behaviour

The dynamics of rainfall, runoff and conductivity at V gauging station, from April to October 2003, are shown Figure 2. The dynamics of total phosphorus, total nitrogen and faecal bacteria are shown Figure 3.

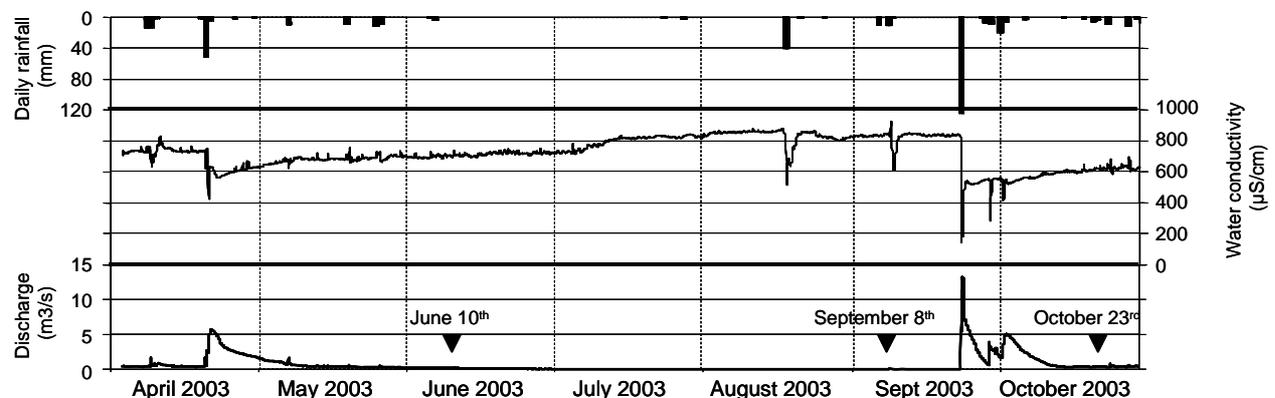


Figure 2 Rainfall, water conductivity and discharge at the outlet (April to October 2003)

During the observation period, a total amount of 405 mm of rainfall was recorded; the total runoff at the outlet V was less than 158 mm. During spring, some rain events of short duration and low

intensity, lead to a succession of small floods. The water conductivities decrease at the beginning of floods, due to surface runoff ($<200 \mu\text{S}/\text{cm}$). The size and the duration of the floods depend on whether the karstic springs are flowing or not, as shown by the water conductivities that are close to those of karstic waters ($\approx 550 \mu\text{S}/\text{cm}$) during long-duration floods. In the late spring, the river discharge become lower than 60 L/s and water conductivity increase slowly. In summer the riverbed falls completely dry, except in some sections where shallow pools remain due to the discharge of waste waters. The discharge at the outlet is just equal to the sewage treatment plant inflow and water conductivities remain constant at the waste water's high value ($>900 \mu\text{S}/\text{cm}$), except during the intense summer rainfalls. The first autumn flood is observed at the end of September, due to a short duration intense rainfall event.

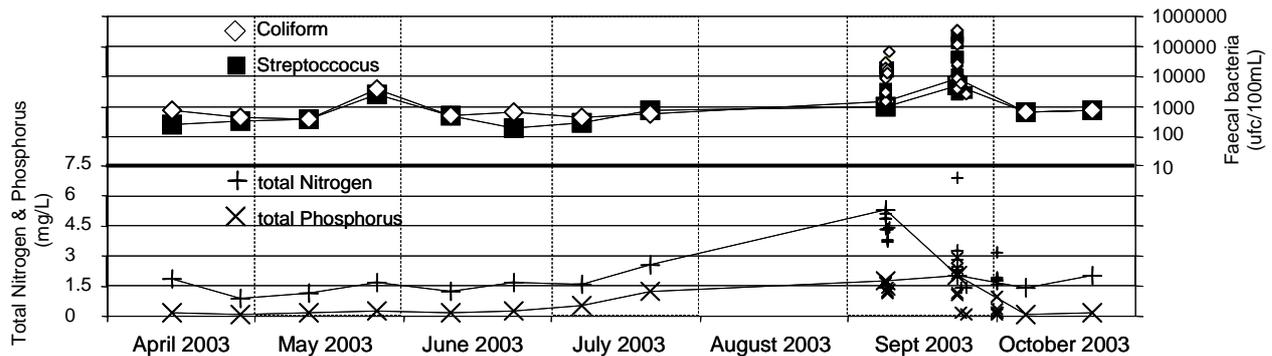


Figure 3 Nutrient and faecal bacteria at the outlet (April to October 2003)

The routine monitoring shows low concentrations of nutrients and high abundances of faecal bacteria. Total nitrogen and total phosphorus concentrations do not exceed 7 mg/L and 3 mg/L , they increase during summer and decrease after the first rainfall events. After the autumn floods concentrations of nutrients recover the spring values. For coliform or streptococcus, concentrations are never lower than $200 \text{ cfu}/100\text{mL}$ and often higher than $1000 \text{ cfu}/100\text{mL}$, especially for samples collected just after a rainfall event.

Flow and concentrations patterns during the campaigns

The one-day campaigns of June, September and October 2003 take place at the beginning of the dry period, just after a small rainfall event and after the first autumn floods, respectively (Figure 2). Figure 4 shows characteristic patterns of the spatial evolution of discharge, concentrations of nutrients and abundances of faecal bacteria along the river. In June, the high spring and the upstream part of the river are dry. With the exception of $T4$, $T5$, $T8$, $T10$ and $T11$, the tributaries are dry and the flow at the outlet is only due to $T10$ and $T11$ inflows: so 80% of the outlet flow is due to the low karstic spring. The pollution levels in the river are low: $\text{TSS} < 45 \text{ mg}\cdot\text{L}^{-1}$; $\text{TN} < 6100 \mu\text{gN}\cdot\text{L}^{-1}$; $\text{TP} < 2200 \mu\text{gP}\cdot\text{L}^{-1}$. The significant increase observed at $R5$ is due to $T5$ sewage treatment plant inflow. Downstream $R5$, the decrease of the concentrations is proportional to the distance. For both nitrogen and phosphorus, dissolved mineral forms represent the main contribution. In September, the river is completely dry, with the exception of large pools of water between $R2$ and $R5$. The discharge at the outlet V is only due to $T10$ inflow. The concentrations of nutrients in the different pools have the same order of magnitude than in June. Organic nitrogen prevails among other species; soluble reactive phosphorus is always over 45% . In October, the river is again (after the large September floods, see here below) in low flow conditions: for the whole river the discharge is lower than 20 L/s ; the discharge at the outlet is explain by the karstic inflow of the low spring plus the inflows of sewage treatment plants. Concentration patterns are very similar to June's one.

In September 22nd, a major rainfall event occurred (125 mm in 7 hours). This event generated a fast and complex flood: the flow at the outlet V which was less than 20 L/s at the beginning of the rainfall event, reached $12 \text{ m}^3/\text{s}$ in 7 hours (Figure 5). The flood can be broken down into three phases.

- The first shower (33.4 mm in 100 min) causes no response at the outlet: the flood was only recorded at the intermediate station S . In fact, runoff volumes are too small for filling the dry course of the

river between *S* and *V*. Conductivity values fall down during the recession: the contribution of surface runoff on urban area is delayed.

- The second shower (92.8 mm in 225 min) causes a generalized flow response. It is worth noting that, in term of volume, there is a limited contribution of the intermediate catchment between *S* and *V*: the low karstic spring is not flowing. Moreover the flood volume is mainly due to surface runoff on the urban areas and the contribution of the agricultural areas is obviously very small, as confirmed by the low water conductivity values at *S* and *V* and the hydrogram recorded at *K* station.
- The last shower (30.4 mm in 125 min) causes a generalized response of the karstic springs: water conductivity values become equal to those of karstic water.

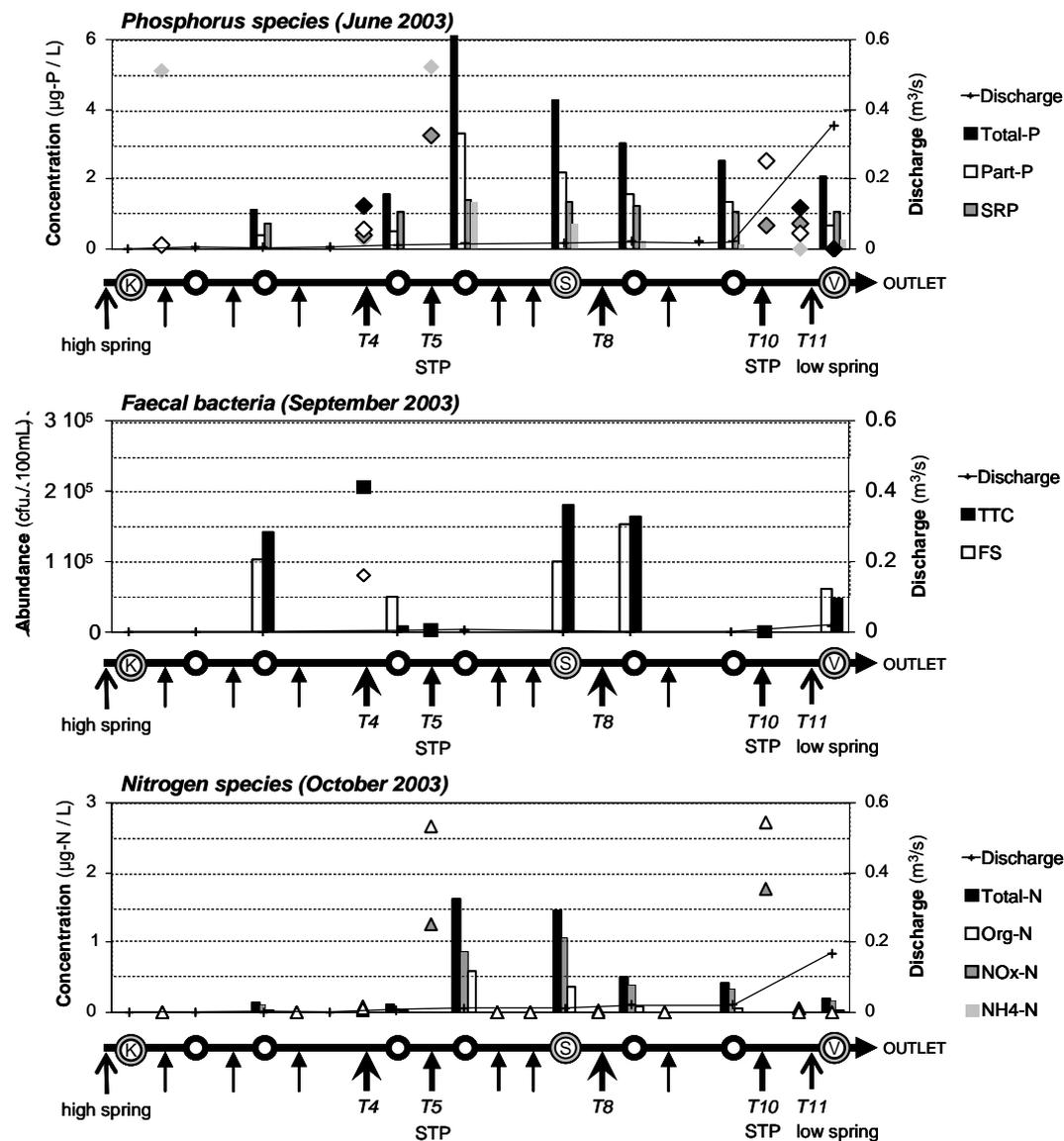


Figure 4 Spatial campaigns : discharge, nutrients and faecal bacteria along the river.

The dynamics of nutrients and faecal bacteria is similar at *S* (Figure 6) and *V* gauging stations (not shown here). At the beginning of the flood, suspended solid are mainly (over 90%) under organic forms, demonstrating that there is a significant flush of the riverbed. The hypothesis is confirmed by the very high values of abundances of faecal bacteria, concentrations of organic nitrogen and total phosphorus. Total nitrogen concentrations show a quick decrease, after the second peak flow. Nitrogen is essentially under organic forms (dissolved and particulate) at the beginning of the flood (over 70 %); nitrates and ammonium show low concentration values along the whole flood. Total

phosphorus, essentially particulate (over 80 %), show the same dynamics as total nitrogen. At the end of the flood, concentrations decrease again, due to the dilution effect by karstic waters.

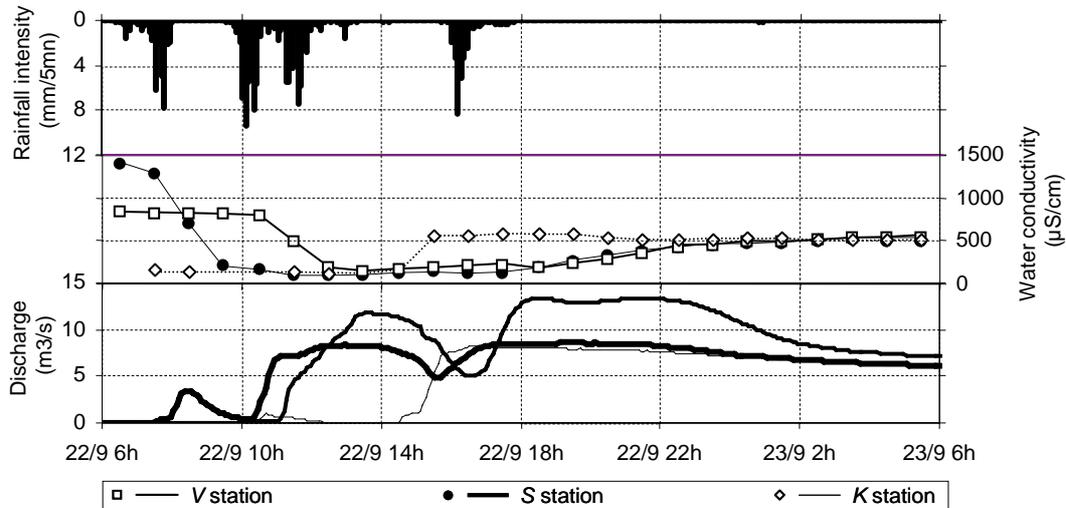


Figure 5 Rainfall, water conductivity and discharge at the 3 gauging stations during September-2003 flood

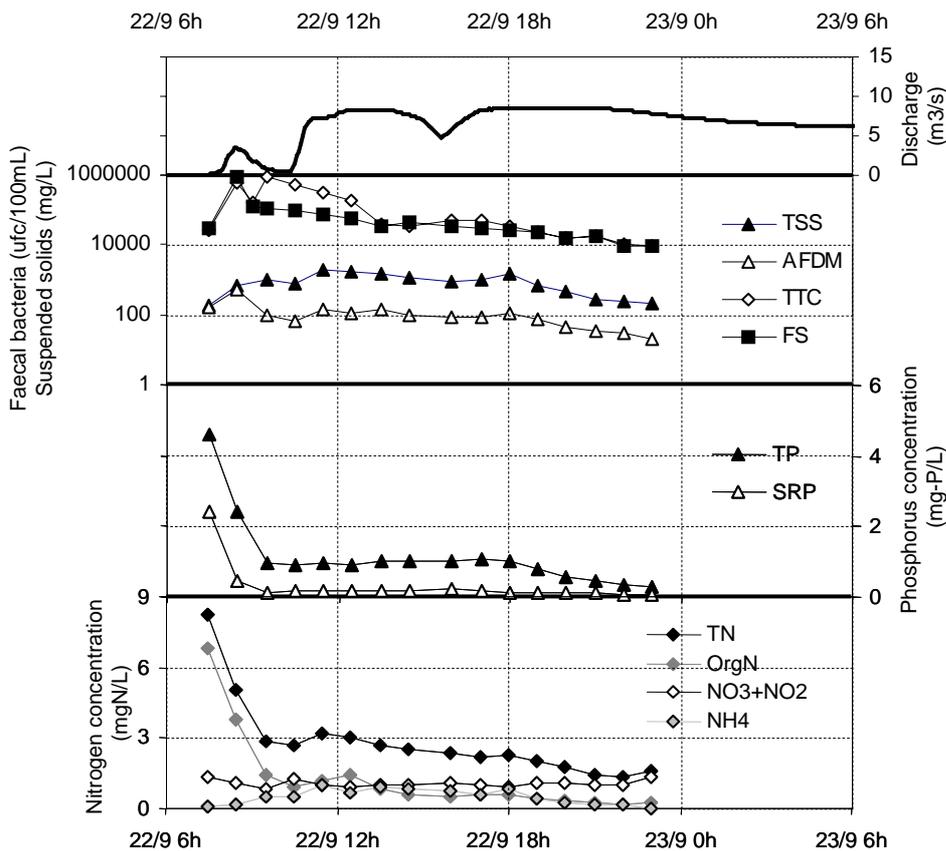


Figure 6 Dynamics of nutrients and faecal bacteria during September-2003 flood (S gauging station)

Evolution of pollutant loads along the river during the summer period

Evolution of loads along the river course is exemplified on Figure 7. In June, the river discharge is mainly due to the inflows of the karstic springs. The sewage treatment plant's inflows represent less than 10% of the discharge. At the contrary, fluxes of nitrogen, phosphorus, suspended solids and faecal bacteria are essentially explained by direct inflows of sewage treatment plant's inflows (80%,

92%, 88%, and over 90%, respectively). This is true for the different species, with the exception of $\text{NO}_x\text{-N}$ fluxes: 69% are brought by the karstic springs, while 6 and 5 % come from soil drainage. But, nutrient and bacteria fluxes showed significant variations along the river. Fluxes decrease along the reaches downstream from the direct inflows ($T5$ and $T10$): from $R4$ to S and from $R7$ to V . Bed sediments played a major role (Tournoud et al., 2005), by trapping TSS and TP in local accumulation reaches. Sedimentation of phosphorus is coupled with co-precipitation of calcium carbonates and sorption in the bed sediments; so, mainly particulate organic forms are retained. The decrease of nitrogen loads along the river is due to bio-assimilation of nitrates and nitrification-volatilisation of ammonium. Sedimentation processes of faecal bacteria are very likely to occur, together with adaptation processes (Sherer et al., 1992). The trapping of nutrients and faecal bacteria is more obvious in September (not shown on Figure 7), since the river is only fed by sewage treatment plants. Large amounts of particulate nitrogen and phosphorus are lost in the pools of waste water remaining few hundred meters downstream $T5$ and $T10$ inflows, from $R4$ to S and from $R7$ to V . The same patterns are observed for October campaign, so the same results can be drawn out since the hydrological conditions are very similar to June's.

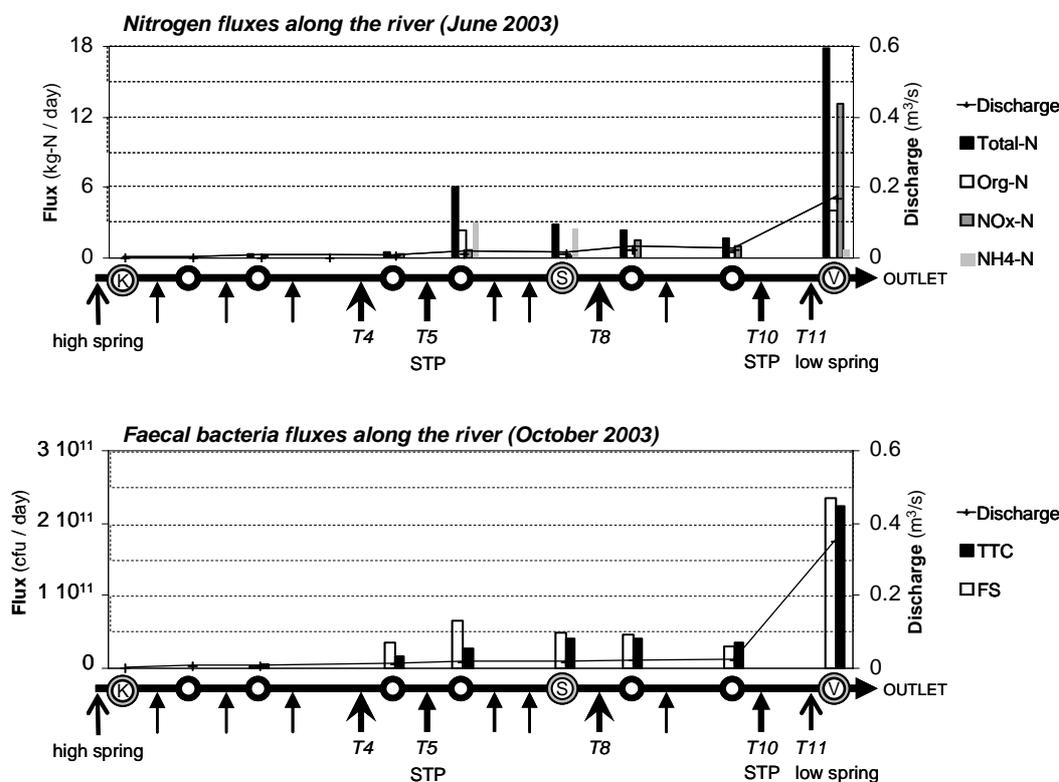


Figure 7 Spatial evolution of loads along the river (examples June 2003 and October 2003)

The mass balances were calculated for the two reaches $R4 - S$ and $R7 - V$, for June and October campaigns, as done by Bowes and House (2001). Then daily retention rates were estimated for typical low flow conditions. The calculation was more hazardous for dry conditions (September), because very low discharges are subject to significant uncertainties. However it was possible to assess the amount of elements that are likely to be retained in the river between the beginning of the low flow period and the first autumn floods, in the river reaches downstream from a direct continuous inflow of waste water. For example, it is estimated that about 250 kg of phosphorus (respectively 300 kg) and 1000 kg of nitrogen (respectively 1700 kg) were retained between $R4$ and S (respectively between $R7$ and V) during the 2003 dry season.

Flush effect of the first flood

The flush effect of the first autumn flood can be highlighted, using a normalized cumulative load plots (Obermann et al., 2007). Figure 8 shows that the ratio of cumulative loads over total flood load is

growing faster than the ratio of cumulative volume over total flood volume: over 75% of the total FS load is flowed since only 40% of the flood volume was flowed through the S gauging stations; the first rainfall event and the associated runoff flowed 30% of the TTC loads, even if it represents only 15% of the total volume. But this flush effect is less significant for dissolved species. Then it can be assessed that the very beginning of the first autumn floods, generated by urban runoff, flushed away the main part of the particulate pollutants, such as faecal bacteria, particulate phosphorus ...

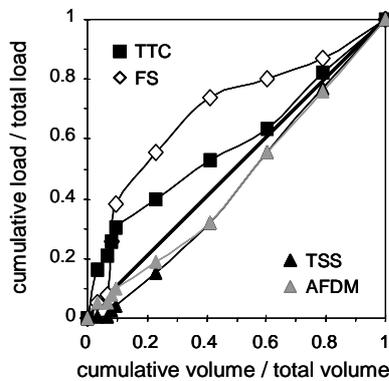


Figure 8 Cumulative load plots (S gauging station)

The comparison of the different origins of nutrient loads shows that about 75 kg of TN is flowed, 20 % coming from rainfall-runoff processes. The rainfall which nitrogen (essentially nitrates) concentrations reach 2000 $\mu\text{g/L}$ brought significant loads at the river, but the washing of urban areas has been assumed, here, to collect no nitrogen. Only 5% of the loads are explained by direct inflows of sewage treatment plants during the first two hours of the flood. So the riverbed itself must be taken into account for explaining the origin of nutrients. It is possible to draw up the same assessment for phosphorus and faecal bacteria.

After all it is possible to estimate that 20 % of the total amount of retained or consumed nitrogen during the summer was flushed away by the flood. But, 50 % of the total amount of retained phosphorus was flushed away at the same time. So the first flood is not likely to exhaust the pollutant reservoir in the river.

CONCLUSION

In intermittent rivers, as it was shown for the Vène River, the low flow period may potentially last several months. It begins very early during spring and lasts even after the first autumn floods. During the low flow period, the river is completely dry, except downstream the continuous inflows from the sewage treatment plants, where pools of waste water remain. Bacteria and chemical elements, continuously discharged, accumulate in these pools, with high organic matter contents. Daily retention rates have been evaluated, for nutrients as well as faecal bacteria.

During the first important rainfall event, in September, a flood was generated mainly by urban runoff. Runoff volumes must be enough to fill up the dry river sections, so that the flood reached the outlet. Important nutrients loads were flushed away during the short duration of the flood.

An important part of these loads probably comes from remobilisation of the pollution retained in the riverbed during the summer. It seems that the stock of pollution is far from being exhausted after the flood. Further work needs to be carried out to validate these assumptions; nitrogen and phosphorus transformation processes must be studied in the accumulation reaches during the summer period; their availability for remobilisation needs to be verified; at last, pollutant stock in the river has to be estimated at the end of the summer period and after the first autumn flood to evaluate more precisely the flush effect of the flood.

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