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Assessment of water pollution in the Antuã River basin (Northwestern Portugal)

M. A. Cerqueira · J. F. Silva · F. P. Magalhães · F. M. Soares · J. J. Pato

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Abstract The Antuã River, located in northwestern Portugal, drains a region with a high population density and a strong economic dynamism. These factors, together with a lack of facilities for appropriate treatment of domestic and industrial sewage, are putting increasing pressure on water resources. In this context, the aim of the present study was to identify point sources of pollution and to assess the surface water quality in the Antuã basin by monitoring physicochemical variables. A total of 40 point sources of wastewater, including some with a high pollution load, were detected in the most populated and industrialized areas of the São João da Madeira and Oliveira de Azeméis municipalities. These sources explained the strong degradation of water quality observed in the upper and medium Antuã River and in one of its tributaries, where maxima of 49 mg l^{-1} for biochemical oxygen demand, 29 mg l^{-1} for Kjeldahl nitrogen and 3.7 mg l^{-1} for total phosphorus, were found after five surface water monitoring campaigns. Despite the relevance of pollution problems, a considerable water quality improvement, promoted by favourable reaeration conditions, was observed in the final stretch of the river, giving evidence of a great self-depuration capacity. However, the Antuã is a significant contributor of nutrients to the Ria de Aveiro, the coastal lagoon where the river meets the Atlantic Ocean.

Keywords Antuã · Monitoring · Pollution · River · Wastewater discharges · Water quality

M. A. Cerqueira (✉) · J. F. Silva ·
F. P. Magalhães · F. M. Soares · J. J. Pato
Centre for Environmental and Marine Studies (CESAM)
and Department of Environment and Planning,
University of Aveiro,
3810-193 Aveiro, Portugal
e-mail: cerqueira@ua.pt

Introduction

The percentage of the European population served by sewage drainage systems has increased regularly since the middle 1980s. Nowadays, the majority of the population from northern and central Europe is connected to wastewater treatment plants that, in many cases, include advanced treatment for nitrogen and phosphorus removal. This change resulted in a decrease in the load of organic matter and nutrients transported by rivers, and subsequently lead to an improvement of the state of water resources (Andersen 2005; Nixon et al. 2003). In Portugal the situation is much less favourable. Two decades after the country joined the European Union (EU), and despite the generous economic support from European funds devoted to investments in sewage systems, only about 60% of the population is connected to public wastewater treatment plants (INE 2005). Under this scenario Portugal was not able to meet the requirements of the Urban Waste Water Treatment Directive (91/271/EEC) – transposed to Portuguese legislation through *Decreto-lei* no. 152/97, *de 19 Junho* (1997)–and is among the group of the former EU 15 Member States with the lowest wastewater treatment rates (EEA 2004). The gravity of the situation is reinforced by the fact that a significant part of the wastewater treatment plants constructed during the last decades present serious operating deficiencies due to, among other reasons, poor project design, lack of qualified staff to operate facilities, and scarcity of investments devoted to maintenance and upgrade programmes.

In this context of fragile sewage drainage and treatment infrastructures, it is not surprising that aquatic resources still reveal degradation signs, as demonstrated in recent studies about the quality of surface waters in Portugal (Cerqueira et al. 2005; Ferreira et al. 2004; Silva et al. 2002).

The Antuã River, located in northwestern Portugal, drains a region with a high population density and a strong economic dynamism. Human settlements in its basin are dispersed over a wide area, giving it a suburban character and making the connection to sewerage infrastructures a difficult task. Adding these factors to the limited capacity of wastewater treatment facilities, results in a great stress on water resources. On the other hand, the Antuã is the second most important river discharging into the Ria de Aveiro coastal lagoon. Thus the pollution load it transports, particularly plant nutrients, has an impact on the Ria ecosystem and is a potential cause of eutrophication processes. In a previous study addressing the eutrophication of the Ria de Aveiro hydrographic basin, Silva and Oliveira (2007) have shown that the Antuã water is enriched with organic matter and nutrients. However the origin of this contamination was not described.

Accordingly, the present study was undertaken to identify and characterize point sources of pollution and to assess the surface water quality of the Antuã basin by monitoring physicochemical variables in selected sampling sites. Whenever possible the water quality status was evaluated using the Portuguese legal framework as a reference, particularly the *Decreto-lei* no. 236/98, *de 1 Agosto* (1998), that establishes quality norms, criteria and objectives to protect the aquatic environment and improve the water quality. This is the first comprehensive study relating the observed water quality with pollution sources generated within the Antuã basin. Information gathered from it increases the understanding of human impacts on the regional aquatic resources and can be an important tool for the authorities in charge of water management, particularly in the context of the new EU Water Framework Directive (2000/60/EC), which requires the implementation of programmes of measures needed to achieve a good ecological status of aquatic systems.

Study area and methods

Description of the study area

The Antuã River arises at Romariz (municipality of Santa Maria da Feira), at an altitude of 400 m, flows for about 38 km, and drains into the Atlantic Ocean, via the Ria de Aveiro coastal lagoon. The corresponding basin, one of the sub-basins of the Vouga River, with a total area of approximately 149 km^2, is oriented in a dominant north-south direction from the source till the site of Minhoteira, where it changes to a west-southwest direction towards the town of Estarreja. The basin includes within its limits parts of seven municipalities: Oliveira de Azeméis, which corresponds to 66% of the basin area, has a central position of prominence, while the others, which share the remaining surface, are displayed on its periphery; São João da Madeira, the smallest of the seven municipalities, with an area of only 8 km^2, is almost totally inserted in the basin; the municipality of Estarreja occupies the lower part of the basin, accounting for 9% of its total area.

The Antuã main tributaries are, from the source to the mouth, on the left margin, the Pintor stream, the Cercal stream and the Ínsua River; and, on the right margin, the Arrifana stream, which drains an important area of the urban centre of São João da Madeira.

The hydrologic regime of the Antuã River reflects the regional expression of climate, which is of the Mediterranean type, with a strong influence from the Atlantic Ocean. Summers are dry and hot and winters are mild and wet, with strong precipitation events. According to the hydrometric records performed near the Minhoteira bridge over the period of 1977 to 1989 (SNIRH 2007), the annual mean flow rate at this point is 4 m^3 s^{-1}, ranging from a minimum of 0.6 m^3 s^{-1} in August to a maximum of 10 m^3 s^{-1} in February.

The Antuã basin underlying geology consists of granite in the upper northeastern reaches, schist in a wide inland area which goes from the source vicinity till the outskirts of Estarreja, and modern alluvial sands and clays in the coastal area that extends from that town till the river mouth. The land use analysis performed by Moreno (2000) revealed that 46% of the basin was occupied by small agricultural fields; 45% with forests of *Pinus pinaster* and *Eucalyptus globulus*; and the remaining area consisted of urban and industrial settlements, dispersed over space in a disorderly manner, due to an accelerated economic growth and an absence of territorial planning.

The total resident population within the basin limits is about 105,000, corresponding to a population density of 705 inhabitants per km^2, a great part of which is present in the territories of São João da Madeira and Oliveira de Azeméis (INE 2002). The percentage of the population from the seven municipalities represented in the basin that is served by sewage drainage systems is lower than 40%, while the percentage of the population connected to wastewater treatment plants varies from 22% in Oliveira de Azeméis to 100% in São João da Madeira (INE 2004a, b). Effluents drained from these two municipalities are sent to a large wastewater treatment plant (WWTP) located at Salgueiro, near the village of Santiago de Riba-Ul. Urban wastewaters from Estarreja are discharged into a regional sewer system, with secondary treatment, that serves 11 municipalities surrounding the Ria de Aveiro lagoon (Silva et al. 2002).

The industrial activity inside the basin limits is dense and diversified, with a predominance of small and medium sized enterprises from the foot-wear, metallurgical, metallomechanical, textile and agro-food sectors. Despite the scarcity of information, the available data suggests that the current situation of industrial drainage systems is quite similar to the previously described for the domestic sector (Moreno 2000).

Animal farming is an important activity in the Antuã River basin, particularly in what concerns dairy cattle management. The majority of the herds are small in size, with cattle production based on semi-intensive management methods in which they are kept indoor for most of the time.

Sampling programme

Field work for the identification and characterization of point sources of pollution (wastewater samples) took place in February and March 2006 in the margins of the Arrifana and Pintor streams and in a stretch of the upper and medium Antuã River that crosses the most populated areas of São João da Madeira and Oliveira de Azeméis.

Campaigns for the assessment of surface water quality were performed on 26 June 2005, 23 July 2005, 20 December 2005, 4 May 2006 and 22 June 2006. The location of the 10 selected sampling sites in the Antuã River basin is shown in Fig. 1. These sites were chosen with the main purpose of identifying critical situations of water quality degradation as a result of pollutant activities as well as to evaluate the river recovery capacity before its outflow in the Ria de Aveiro. Sampling sites designation and its exact location in the basin is as follows: site 1 (Milheirós de Poiares) was situated near the source of the Antuã River and was selected to represent a non-polluted water condition; site 2 (São João da Madeira) was located southwest of the city of São João da Madeira and downstream of a small industrial area; site 3 (Samil) was selected to evaluate the water quality of the Pintor stream, just before its conjunction with the Antuã River; site 4 (Faria de Cima) was located on the Arrifana stream, after the urban settlement and the southern industrial area of São João da Madeira; sites 5 (Ferral) and 6 (Aguincheira) were located on the Antuã River, respectively upstream and downstream of the Salgueiro wastewater treatment plant, in order to observe possible impacts of sewage discharges from that facility in the receiving waters; site 7 (Salgueirinha) was selected to study the influence of domestic and industrial wastewater discharges from the surrounding area; site 8 (Ul) was chosen to monitor the water quality of the Ínsua River; site 9 (Minhoteira) was located after the confluence of the Ínsua River, to evaluate the effect of flux from this tributary on the Antuã waters; and site 10 (Estarreja) was established a few km upstream of the mouth, to monitor the quality of the water discharged into the Ria de Aveiro coastal lagoon. All the samples were collected from bridges, except on site 4, where sampling was performed from the left bank of the Arrifana stream.

Sampling and analytical methods

Sources of pollution with easy and safe access were characterized for flow rate and physicochemical parameters. Flow rate was determined by the measurement of

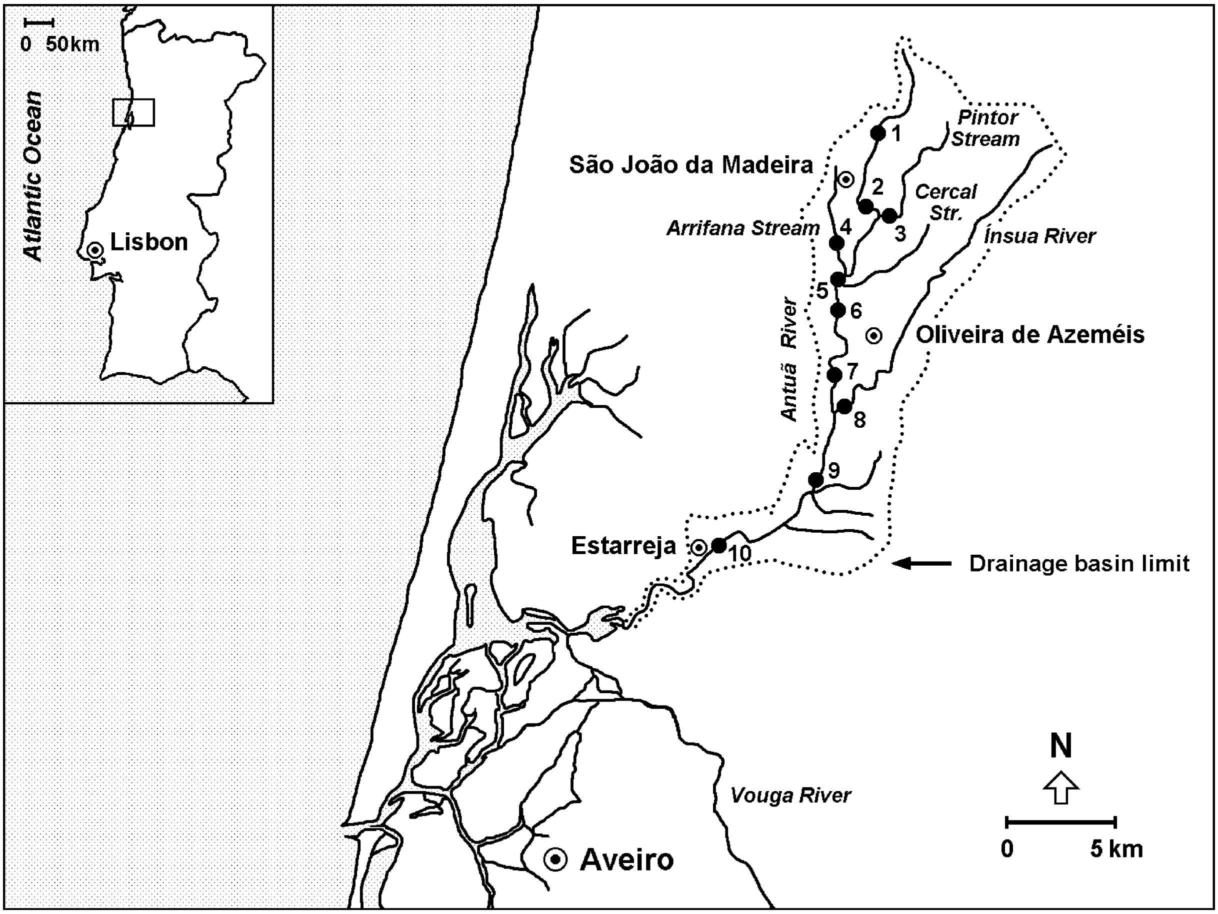

Fig. 1 Map of the Antuã River basin showing the location of sampling sites

time required to fill a bucket of known size. Wastewater samples were collected with 1 l acid-washed polyethylene bottles and transported to the laboratory where they were analysed on arrival for chemical oxygen demand (COD), conductivity and total suspended solids (TSS).

Samples for the assessment of surface water quality were taken at each site using a bottom weighted polyethylene flask, previously washed with water from that site. Temperature, dissolved oxygen and conductivity were measured in the field with portable meters. The samples were then transferred to 2.5 l acid-washed polyethylene bottles, transported to the laboratory and processed within 6 hours after collection for the analysis of pH, TSS and biochemical oxygen demand (BOD_5). Aliquots of the initial samples were also preserved for later analysis of Kjeldahl nitrogen (Kjeldahl-N), nitrate and total phosphorus. TSS were assayed by gravimetry after filtration of an adequate sample volume through a glass fibre filter and drying at 105°C. BOD_5 was determined as the difference between initial and 5-day dissolved oxygen content, after incubation in the dark at 20°C. COD was measured by using the dichromate open reflux method. Kjeldahl-N was measured by digestion and distillation, according to the Kjeldahl method, and subsequent analysis of ammonia by the colorimetric indophenol blue method. Total phosphorus was measured as orthophosphate by the colorimetric ascorbic acid method after acid digestion. Nitrate was determined by ion chromatography with suppression. Data quality was guaranteed through the implementation of laboratory quality assurance and quality control methods, including the use of standard operating procedures, calibration with standards, analysis of reagent blanks, recovery of known additions and analysis of replicates. All the analytical protocols used during this study were in accordance with the standard methods described in APHA (1995).

Results and discussion

Hydrological conditions

Figure 2 shows rainfall data from the meteorological station of Castelo de Burgães (located ≈10 km east of Oliveira de Azeméis) along with the Antuã River flow recorded at Estarreja for a period that covers field surveys performed as part of this study. Flow measurements started only in May 2005 in the framework of a water resources monitoring programme in progress at the lower Vouga River basin. The first months of 2005 were characterized by severe drought conditions that affected the whole country. Rainfall from January to June of 2005 was 238 mm, corresponding to only 30% of the 1990–99 average. In the following 6-month period the accumulated rainfall was 469 mm, and the year of 2006 brought wetter conditions, with rainfall returning closer to normal values (669 mm from January to June). The seasonal variation of flow rate reflected the trend of rainfall, as the lowest monthly mean values were recorded in the summer of 2005 and the highest during the spring of 2006.

Wastewater discharges

A significant number of point sources of pollution (40 in total) discharging wastewater into the Antuã River or its tributaries was identified during the field surveys performed in the municipalities of São João da Madeira and Oliveira de Azeméis. The major part of these sources was the result of direct discharges from domestic sewage drainage systems not linked to any treatment facility or from improperly operated or poorly maintained septic tanks.

Sewage characterization was possible only for 20 of the above mentioned point sources. The corresponding results are provided in Table 1.

The flow rate of wastewater discharges at the moment of sampling varied over five orders of magnitude. Only one half of the monitored sources exhibited a flow rate above 3 $m^3\ h^{-1}$ and the two highest values (72 and 342 $m^3\ h^{-1}$) were recorded in effluents derived from a milk processing factory and from the Salgueiro WWTP, respectively. Together, the main sources of pollution amount to more than 500 $m^3\ h^{-1}$ of liquid waste released into the Antuã, a significant value since the average river discharge is low, particularly in summer.

Measurements of COD, conductivity and TSS demonstrated that some of the monitored effluents have the typical characteristics of domestic sewage found at European sites (Pons et al. 2004). However the mean values are indicative of a weaker pollution load for the majority of the effluents. These results may be explained by the fact that most samples were collected from wastewater released by septic tanks, where a certain degree of treatment is expected to occur. Another possible explanation is the infiltration and inflow of water into the sewer systems, which may have been relevant during the wet period of February and March of 2006. Samples collected from the two main sources of pollution (in terms of volume)

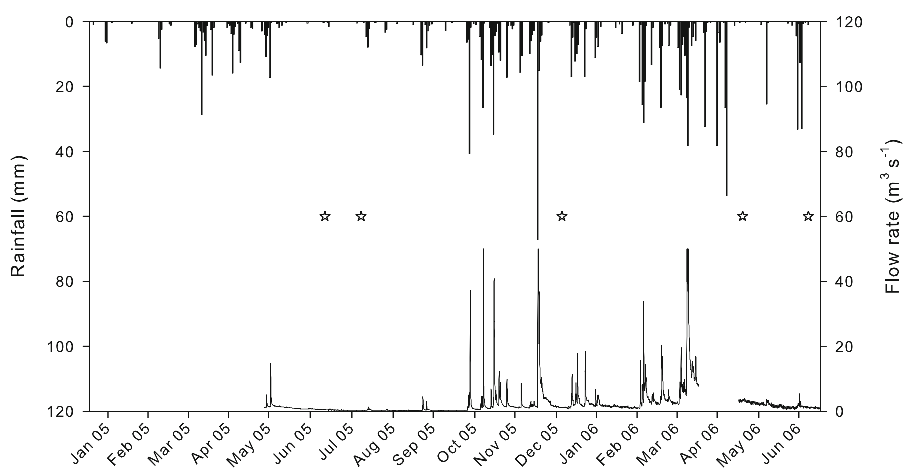

Fig. 2 Temporal variation of rainfall at Castelo de Burgães (SNIRH 2007) and flow of the Antuã River at Estarreja for the period of January 2005 to June 2006. The *stars* indicate the time of surface water monitoring

Table 1 Statistical data of flow rate and physicochemical properties for the point sources of wastewater characterised in this study

	Flow (m^3 h^{-1})	COD (mg l^{-1})	Conductivity (μs cm^{-1})	TSS (mg l^{-1})
Average	43.5	203	760	62
Median	3.4	316	987	68
Minimum	0.03	35	414	12
Maximum	342	1039	1717	715

found during the field campaigns revealed concentrations below the legal limit for COD and TSS in effluents (*Decreto-Lei* no. 236/98). Nevertheless, their impact on the water quality of the Antuã River cannot be ignored. This is particularly relevant for the Salgueiro WWTP, which was discharging into the river a COD load of 16 kg h^{-1}, about one third of the total load released by all the monitored pollution sources.

Surface water quality

The spatial and temporal variations of the monitored parameters in the surface waters of the Antuã River basin are shown in Figs. 3 to 5.

Temperature values were in the range of 15.8 to 21.9°C during the summer campaigns and 7.1 to 11.2°C during the winter campaign. The spatial variation followed a common pattern for most of the sampling events: minima were usually observed near the source and in tributaries draining the upper basin lands (to the east), and maxima were recorded in the lower stretch of the Antuã River. These trends were not surprising and can be attributed to altitude differences of sites and climate seasonal variation. A somewhat different spatial variation was observed in December, when the water temperature remained quite constant along the Antuã River and the highest temperature value was detected in the Arrifana stream (site 4), and in May, with a peak in São João da Madeira (site 2). These deviations from the common temperature profile suggest an overheating of surface waters due to warmer wastewater discharges in the basin northwestern area, where most of the population and industries are concentrated.

The temporal variation for conductivity was quite similar in the majority of the sampling sites: an increase from June to July, a decrease in December, a minimum in May and then a rise in June of 2006. These results are a consequence of variations in the concentration of dissolved solids due to the seasonal fluctuations in the amount of water drained by the hydrologic system. The minimum conductivity values were recorded at site 1, near the source of the Antuã River, and at site 8, in the Ínsua River, where the impact of pollution sources is of scarce relevance. Maximum values occurred at sampling sites 4, 6 and 7, and can be attributed to the high concentration of point sources of pollution found in this area. The conductivity values at Minhoteira (site 9) were lower than at Salgueirinha (site 7) by a factor of 1.4 to 1.7, reflecting the dilution effect that resulted from the discharge of the Ínsua tributary into the Antuã waters.

The pH results were very uniform throughout this study, with minor spatial and seasonal differences. All the samples exhibited values in the range of 6.3 to 7.7 units of pH, except those collected at Samil (site 3), in May and June 2006, which were a little more acidic, seeming to reflect the impact of acid waters draining from the gallery of an old arsenic mine located in the right margin of the Pintor Stream. According to a previous field work, performed in the summer of 1996, the stream waters experienced a pH reduction of 2 to 3 units at a site just after the mine effluent discharge, about 2 km upstream of Samil (Moreno 2000). Comparing pH results from the present study with the Portuguese legislation (*Decreto-Lei* no. 236/98) it is possible to notice that all the values fall within the limits established as an objective of minimum quality for surface waters (5.0–9.0).

The lowest concentrations of TSS were recorded near the headwaters of the Antuã River (site 1) and in the lower Ínsua River (site 8), since upstream of these sites both water courses drain areas on which the anthropic disturbance is minimal. TSS levels were also low in samples collected from site 5, but increased 2 to 40 times in the downstream station (site 6), just after the Salgueiro WWTP, a facility with a capacity already exceeded, releasing frequently non-treated effluents into the adjacent river. Another increase was observed further downstream, at Salgueirinha (site 7), reflecting the impact of a large number of sewage discharges, particularly from the domestic sector. In fact, collection sewers serve the nearby villages of Santiago de Riba-Ul and Ul but wastewater gathered from local users is transported directly into the river, because the main sewers are not yet linked to any treatment facility. The TSS value of

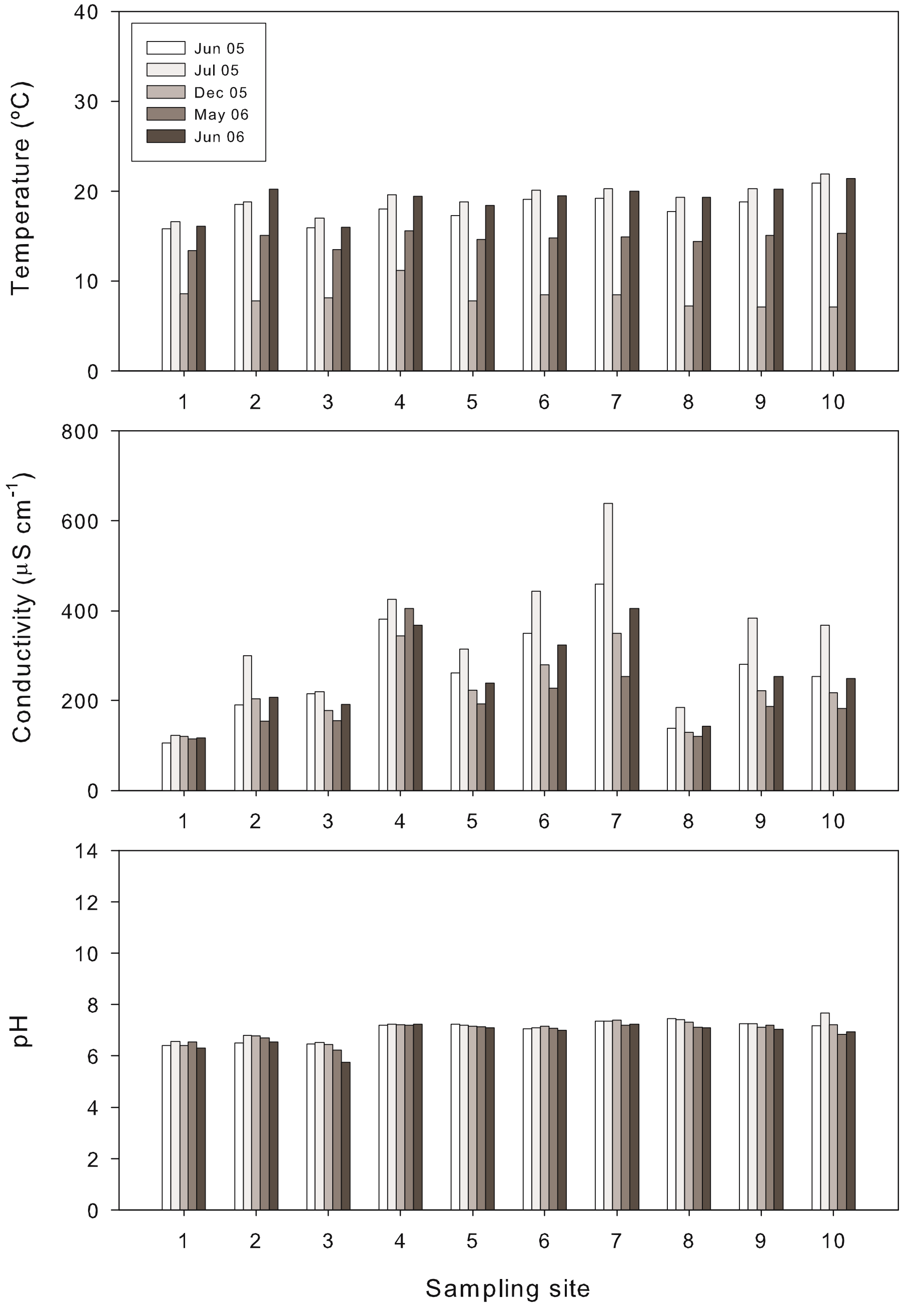

Fig. 3 Spatial and temporal variations of temperature, conductivity and pH in the surface waters of the Antuã River basin

42 mg l^{-1} that was recorded at site 2 in December was in clear disagreement with results from the other campaigns and may attributed to riverbank rehabilitation works that were in progress in the area of São João da Madeira. A regulatory level was not defined in the Portuguese legislation for TSS in surface waters.

As can be seen from Fig. 4, the basin surface waters appeared to be fairly well oxygenated. The only exceptions were sampling sites 4, in the Arrifana stream, and sites 6 and 7, in the Antuã River, where the percentages of saturation for the summer of 2005 were lower than the objective of minimum quality for surface waters prescribed by the Portuguese law (50%). These values were linked to a very high load of organic matter – which is known to consume oxygen when it decays. BOD_5 results for those three sites clearly stand out from the set of measurements performed in the basin, varying between 9 and 49 mg l^{-1}, well above the legal limit of 5 mg l^{-1}. Results from the Arrifana stream (site 4) were attributed to non-treated wastewater discharges with an origin in the urban settlement and in the southern industrial area of São João da Madeira. On the other hand, results from the Antuã River (sites 6 and 7) were related to the above mentioned operating conditions of the Salgueiro WWTP, to domestic effluent discharges from the nearby villages and to sewage coming from local industries. A considerable improvement on the water quality was observed going downstream, towards Estarreja (sites 9 and 10). This was promoted by a series of weirs located in the medium stretch of the Antuã, used in the past to create mill ponds, which generates waterfalls and adds oxygen to the river. The water purification was also facilitated by the dilution effect that resulted from the confluence of

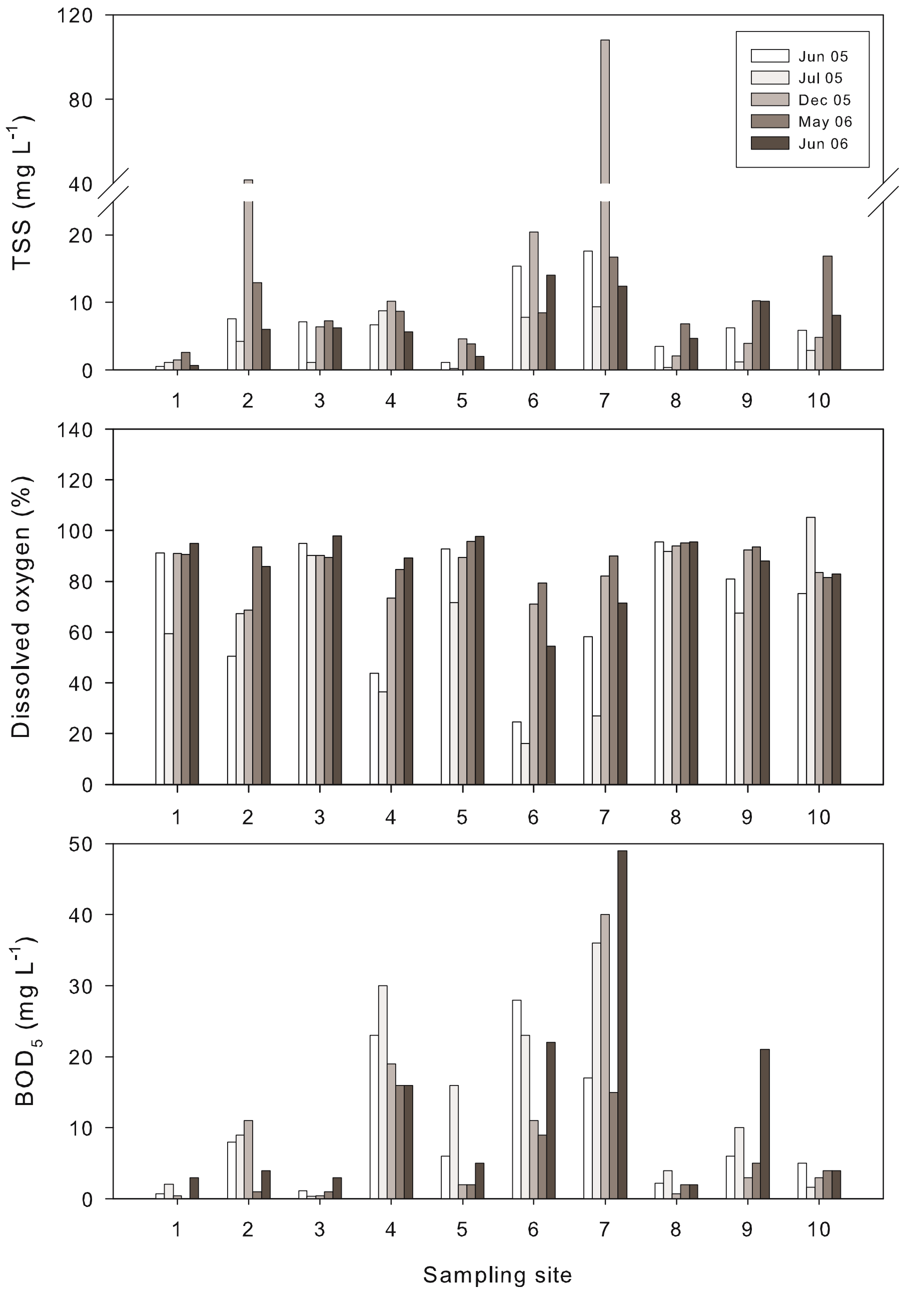

Fig. 4 Spatial and temporal variations of total suspended solids, dissolved oxygen, and BOD_5 in the surface waters of the Antuã River basin

the Ínsua River, which possessed a low organic load (BOD_5<5 mg l^{-1}), with the Antuã River.

The highest Kjeldahl-N values were also observed at sites 4, 6 and 7, whatever the sampling campaign, with concentrations in the range of 3 to 29 mg l^{-1}, in excess of the Portuguese legal standard of 2 mg l^{-1}. On the other hand, minimum values were measured near the source of the Antuã River (site 1), in the Pintor stream (site 3) and in the Ínsua River (site 8). Considering that high levels of reduced nitrogen compounds are a sign of inadequate sanitary conditions due to a fresh contamination with organic wastes (Sawyer et al. 1994), these results demonstrate that sewage discharges from urban areas are strongly affecting the Arrifana stream as well as the stretch of the Antuã River that runs west of Oliveira de Azeméis.

An opposite spatial pattern was observed for nitrate. Sites 4, 6 and 7 were now included in the group of places with the lowest concentration values, while sites 9 and 10, in the lower course of the Antuã, exhibited some of the highest nitrate levels. This trend shows that nitrogen reduced forms tend to be dominant under conditions of greater organic pollution load and lower oxygen content, like those prevailing near the urban areas of São João da Madeira and Oliveira de Azeméis in the summer of 2005, and then undergo a gradual oxidation to nitrate when the water flows downstream and dissolved oxygen increases by reaeration. However, only part of the Kjeldahl-N load was transformed and accumulated as nitrate. A simple mass balance reveals that about 2/3 of this load is removed from the water, bringing to evidence the importance of ammonia volatilization and denitrification for the nitrogen cycling. Analysis of the data in Fig. 5 also indicates moderate nitrate concentrations at Milheirós de Poiares (site 1) and Ul (site 8). Considering the limited number of point sources of pollution in areas located upstream of these sites, the observed levels are indicative of nitrate inputs from diffuse agricultural sources. A legal limit was not defined in the Portuguese law for nitrate in surface waters.

With regard to total phosphorus, once again the Arrifana stream (site 4) and the part of the Antuã River that flows west of Oliveira de Azeméis (sites 6 and 7) revealed the highest concentrations, most of the times near or above the legal limit of 1 mg l^{-1} established by the Portuguese law, which were certainly linked to the above mentioned point sources of pollution. Enhanced phosphorus concentrations are common in surface waters that suffer the impact of urban wastewater discharges, as a result of domestic use of detergents containing phosphates (e.g. Jordão et al. 2007; Kannel et al. 2007). Interesting to note is the lower decrease of phosphorus levels going downstream to the river mouth, particularly when compared with the spatial variation of Kjeldahl-N. This trend shows that phosphorus was not transformed or easily assimilated by aquatic vegetation. Thus, the dilution effect that resulted from the discharge of the Ínsua waters was a dominant factor explaining the declining concentrations between sampling sites 7 and 10.

Conclusions

The monitoring campaigns performed as part of this study demonstrated that the upper and medium course of the Antuã River receives liquid wastes from various point sources of pollution located in the municipalities of São João da Madeira and Oliveira de Azeméis. Most of these sources are the result of direct discharges from small domestic sewage drainage systems not linked to any treatment facility or from improperly operated or poorly maintained septic tanks. The main source of pollution in the area is the Salgueiro WWTP, a large facility that receives sewage from three municipalities and does not provide an adequate treatment capacity. The impact of effluent discharges was evident in the Arrifana stream and in a stretch of the Antuã River that crosses Oliveira de Azeméis, where the measurements of BOD_5, Kjeldahl-N and total phosphorus revealed values well above the minimum quality objectives for surface waters set by the Portuguese legislation. No significant differences were detected in the quality of water on a seasonal basis, but the combined effect of drought conditions and warm ambient temperatures seems to induce high oxygen consumption in areas strongly affected by effluent discharges. Despite the relevance of pollution problems in the Antuã, a considerable water quality improvement, promoted by favourable reaeration conditions, was observed in the final stretch of the river, giving evidence of a great self-depuration capacity. Nevertheless, the concentration of nutrients near the river mouth was high enough to be a cause

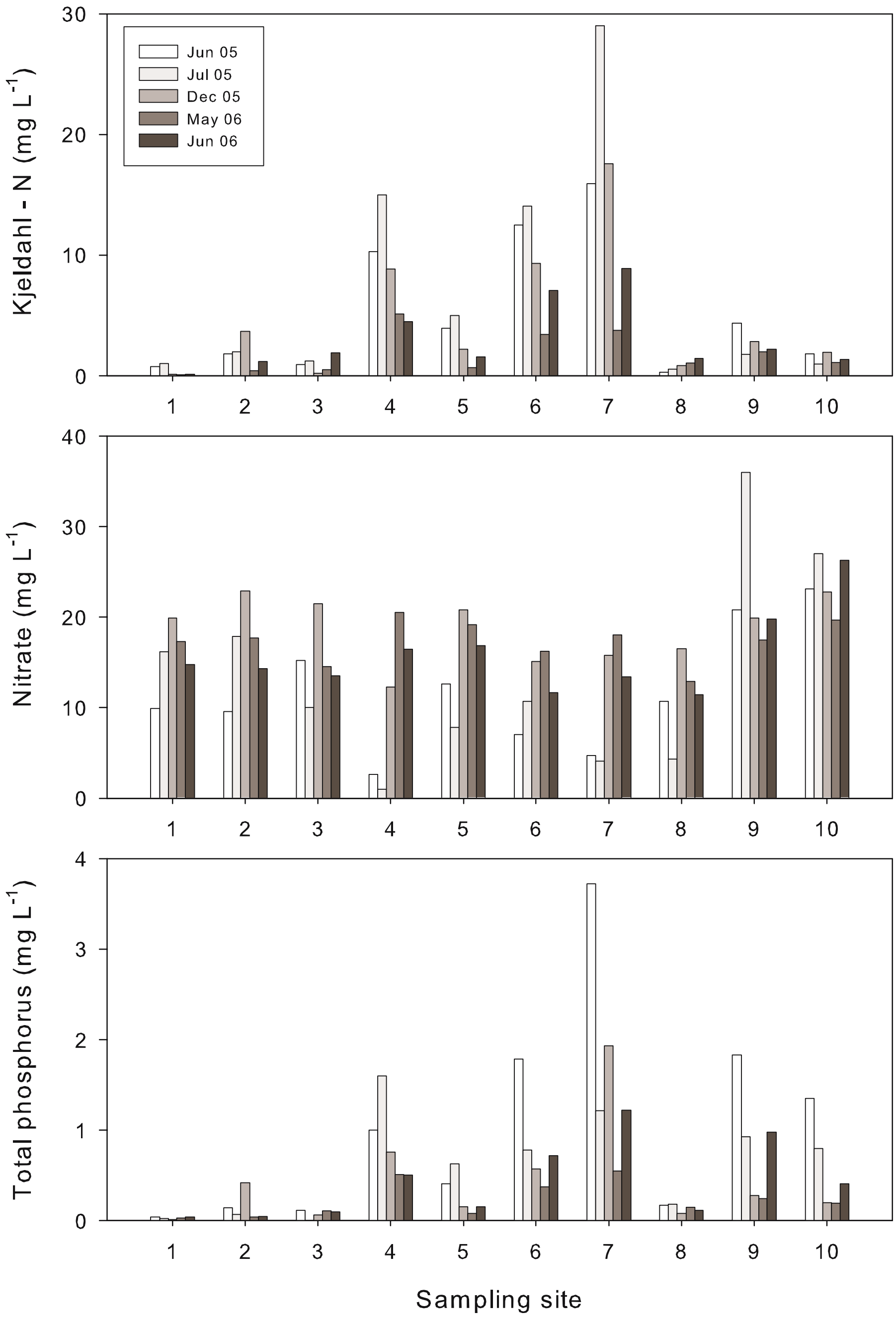

Fig. 5 Spatial and temporal variations of Kjeldahl-N, nitrate and total phosphorus in the surface waters of the Antuã River basin

for concern about possible impacts in the Ria de Aveiro coastal lagoon.

Finally, results presented in this study emphasize the need for additional efforts directed towards the protection of water resources. Priority tasks should include the control of point source discharges by means of greater sewage network coverage and improved capacity of wastewater treatment facilities. But, any action in this field cannot be separated from the development and application of efficient territorial planning instruments, in order to prevent the current expansion of residential and industrial areas in a scattered and disorderly manner which renders it difficult to control pollution sources and promotes wasting of resources.

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