The complex task of maintaining water quality in Mediterranean basins: Case study, Llobregat River basin, Spain

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Abstract Many river basins in Southern European (SE) countries suffer common freshwater quality and management problems. Although in recent decades there has been an improvement in wastewater treatment and in the number of people connected to treatment facilities, the eutrophication problem has not decreased, and emerging contaminants are becoming a new social concern. The irregular Mediterranean hydrology, wastewater discharges from highly populated areas and the extensive use of water resources for agriculture are the main environmental pressures of Mediterranean basins where maintaining adequate water quality, and good environmental status, while providing a reliable water supply, is a difficult task. In many of these basins water scarcity and long drought periods affect river flows and, consequently, the dilution and self-purification capacity of water bodies. Furthermore, diffuse and groundwater pollution by agriculture, and the massive use of water resources by irrigated agriculture are other main factors in river and aquifer water quality. However, new water policies, including the European Framework Directive, are demanding more environmental protection for our water bodies. In this paper we explain the common stressors of the environment and water quality in these southern countries' basins. As an example of such basins we present the Llobregat River basin (LRB) case, in Spain. The importance of this river is that it is the main water supply resource of several cities, including Barcelona, and the main receptor of effects of different human activities. Water quality is compromised by human environmental pressures and low river flows in scarcity periods.

Key words water quality; water quantity; Southern European countries; Mediterranean basins; Llobregat River, Spain

INTRODUCTION

Water quality is a key factor in water use in many basins around the world. The relationship between water quantity and quality is twofold. On the one hand, the increase of water use has caused the reduction of river flows, less dilution and reduced self-purification capacity. On the other hand, water quality is a more important restriction to water resources uses every day. Forecasts of population, food needs and climate change predict an increase of water quality problems in the near future.

Water quality in Europe is drastically different from one country to another, depending on many physical and socio-economic factors. However, the Southern European (SE) countries are subject to many environmental pressures obstructing the achievement of the good ecological status required by European Water Framework Directive (WFD, EC 2000). The WFD requires that in all member states all water bodies achieve good ecological status by 2015. This good ecological status is defined by various biological, physico-chemical and hydromorphologic conditions. Although recently published water basin plans show that hydromorphology and alteration of habitats are the most common pressures, physico-chemical problems are probably the main social, economic and continuing problem across the European Union (EU).

This paper presents the main water quality problems and drivers of continental water in SE countries with a focus on Mediterranean basins. Although in many studies and reports environmental data and analysis in Europe are classified by country, in SE countries water quality issues are exacerbated differently from one basin to another. Consequently, it is necessary to remark on aspects of water quality in Mediterranean basins. We show that in these countries water quality is closely bound to water quantity, and that, in order to achieve good water quality status of water bodies, water management is at least as important as water treatment. Finally, we present the current water quality situation of the Llobregat River basin, in Spain, as an extreme case of the problems presented. The Llobregat River basin presents a high use of water resources while water quality problems arise in its middle-lower part. The management of the basin to supply all water demands and maintain water quality is an extremely complex task.

WATER QUALITY STATUS IN SOUTHERN EUROPEAN COUNTRIES AND MEDITERRANEAN BASINS

Providing an explanation of water quality in SE countries is a complex task due to several factors. First, compilations and databases are at national scale without distinctions between basins or sub regions. SE countries present marked differences from north and south or, as occurs in Spain, between the Atlantic and Mediterranean watersheds. Precipitation, air temperature, topography, soil and other physical features present high spatial gradients in these countries. Consequently, some water quality and quantity key factors present many differences between different areas. For example, in Spain mean annual precipitation ranges from 1200 mm/year to less than 200 mm/year. A second factor is the general lack of data in reviews and reports from the EU. For example, the Environmental European Agency (EEA) 2003 report claims that, at present, it is not possible to obtain an overview of the ecological status of European waters as there are many significant shortfalls and gaps in information, monitoring and assessment systems from the different estate members. Finally, SE countries have evolved differently in the last decades in terms of wastewater treatment investments.

Even with this lack of data, herein we present a summary of several key factors and drivers of water quality in SE countries based on scientific references, on EEA reports, and on our experience on water quality analysis and modelling in several Spanish river basins (Paredes *et al*., 2010; Paredes-Arquiola *et al*., 2010).

At European level, some improvements in water quality were made in the last two decades with the application of the different European water quality directives finally embraced in the WFD. However, the current ecological status of water ecosystems is not good enough. The content of oxygen consuming substances in rivers has decreased in European rivers in the last decades (EEA, 2012c). In the case of the SE rivers (represented in this report by the Spanish rivers only) they reached the lowest level in 2003. From our experience with Spanish data, we can conclude that in the 1990s and the first decade of the 21st century, water treatment investments were focused on secondary treatments and a progressive augmentation of people connected to wastewater treatment facilities. This brought a reduction of BOD_5 in many water bodies. But the ammonia concentrations have remained similar, or even have increased in some places. In many Spanish rivers the main process of consuming dissolved oxygen has changed from degradation of organic carbon to nitrification of ammonia. Although the mentioned EEA report presents a drastic diminution of ammonia concentrations in rivers in the last decades, they specify that for Southern countries only Spanish data are available and these present the lowest decrease (20%) compared to other countries. Finally, it is remarkable that in recent years, mainly since 2005, there has been an increase of tertiary treatments reducing nitrogen and phosphorous. This has been reflected in river concentrations, including an improvement in dissolved oxygen concentrations. A collateral effect is an increase of nitrates concentrations just downstream of wastewater treatment plant (WWTP) discharges. This evolution of Spanish WWTP could be translated to other SE countries, but not all of them. The rate of population connected to wastewater treatment is different in each country and while in Greece or Spain this is over 80%, in other SE countries it presents values lower than 20% (EEA, 2012c).

Together with organic matter, nutrient pollution is another general water quality problem. Excessive concentrations of phosphorus are the most common cause of freshwater eutrophication (Correll, 1998) and source apportionment studies indicate that, in general terms, agriculture and point discharges are the key sources of phosphorus found in European freshwater bodies (EEA, 2005, 2010). Until now, in SE countries, with less tertiary treatments than Northern countries, urban and industrial discharges were the main sources of phosphorous. Recent wastewater treatment improvements are notably reducing concentrations in rivers and lakes/reservoirs, but it is still a generalized problem.

However, the main source of nitrates is agricultural activity, especially in irrigated areas. The problem of nitrates is generalized across European countries. Diffuse pressures are largely driven by nitrates, mainly coming from fertilizers, which runoff into water bodies. If the current trend continues, concentrations of nitrates in water are unlikely to meet good status values within the next 10 to 15 years (EEA, 2012b). Mediterranean regions present major problems of high nitrate concentrations in surface and groundwater. For example, groundwater nitrate pollution in the Po valley (Italy) has been widely studied.

Irrigated agriculture causes various direct or indirect problems, including leaching of nutrient and pesticides, soil salinization, overexploitation of aquifers (leading to ground subsidence and seawater intrusion), modification of natural flow regimes and damage to water dependent ecosystems (Wriedt *et al*., 2009). Despite improvements in some regions, diffuse pollution from agriculture remains a major cause of the poor water quality currently observed in different parts of Europe (EEA, 2010). Source apportionment studies indicate that, generally, agriculture contributes to 50–80 % of the total nitrogen load observed in Europe's freshwater, with point discharges, including from wastewater treatment plants, providing much of the remainder (EEA, 2005, 2010).

Furthermore, new and largely unknown groups of substances keep appearing in the aquatic environment. The effects of these substances may be even more significant than more traditional pollutants. Some examples are antibiotics, medicines and substances that disrupt hormonal balance in humans and animals (EEA, 2012b). European chemical and associated industries have developed rapidly in recent decades, making a significant contribution to the economy and to the global trade in chemicals (EEA, 2011). A number of recent European research studies have contributed to a growing, although still incomplete, understanding of the levels of various hazardous substances in water, including some of those viewed as emerging pollutants (EEA, 2011). A group of these chemicals are known as Endocrine Disrupting Chemicals (EDC). Sewage treatment plants can remove 80% or more of them (Baronti *et al*., 2000; Andersen *et al*., 2003), resulting in low concentrations – around tens of nanograms per litre – subsequently being discharged to receiving waters. However, even such low concentrations can result in endocrine disruption in aquatic biota (Jobling *et al*., 2006; EEA, 2010). These studies include surveys of pharmaceuticals in freshwater in Spain (Ginebreda *et al*., 2010) and the UK (Ashton *et al*., 2004) and heavy metals in the Seine (Meybeck *et al*., 2007). In Spain, vitellogenin was found to have increased in male carp downstream of a sewage treatment plant (Petrovic *et al*., 2002) whilst in Italy, 50% of barbel sampled below a polluted tributary of the River Po showed intersex gonads (Viganò *et al*., 2001; EEA, 2011).

Water exploitation

Different indexes have been used to define the pressure or stress on freshwater resources. The Water Exploitation Index represents the ratio of total freshwater abstractions to the total renewable resource (EEA, 2009). According to Raskin *et al*. (1997) a WEI above 20% implies that a water resource is under stress, and above 40% indicates severe water stress. With the annual estimation Cyprus shows the highest value (45%) and other Mediterranean countries such as Spain (34%) present similar severe values of stress at the national level (EEA, 2009). However, the same index estimated by region shows that the Andalusia region and the Segura River basin (south and southeast of Spain) have extreme values of 164% and 127%, respectively. For example Pinios RBD in Greece is severely water stressed. Nevertheless this is compensated at the country level WEI. Similarly, the regional WEI in Portugal is extremely variable per river basin (Sado RB 132%, Leca RB 82%, Minho RB 1%, Lima RB 5%) but at country level is averaged at 15% (EEA, 2012c). Furthermore, these assessments are considered at the annual scale not taking into account the effect of the concentration of the demand in the summer months.

More data that confirm the water use of SE countries, and especially in Spain and Turkey, show that the former is the country with the greatest number of reservoirs in the EU, and both countries are able to store more than 40% of their renewable resource (EEA, 2009). Moreover, Spain is the largest user of desalination technologies in the western world (EEA, 2009). Other Mediterranean countries, e.g. Cyprus, Greece, Italy, Malta and Portugal, also rely increasingly on desalinated water as an additional resource for public water supply and to support holiday resorts in arid areas (EEA, 2009). Finally, Spain and Italy are the countries with the greatest re-use of water in the EU. From 964 hm^3/year of water re-used in the whole EU, Spain and Italy re-use 347 and 233 hm^3/year, respectively (Mediterranean EUWI Wastewater Reuse Working Group, 2007; EEA, 2009).

According to the same report, the total abstraction of freshwater across Europe is around 288 km^3/year and represents, on average, 500 m^3 per capita/year. Overall, 44% of the total abstracted is for energy production, 24% for agriculture, 21% for the public water supply and 11% for industry, although strong regional variations are apparent. In SE countries, the largest abstraction of water is for agricultural purposes, specifically irrigation, which typically accounts for about 60% of the total abstracted, rising to 80% in certain locations (EEA, 2009). In SE, irrigated agriculture is by far the largest consumer of freshwater resources (Wriedt *et al*., 2009). Agriculture is an essential driving force in the management of water use having significant impacts on water quantity and water quality. This is especially true in the Mediterranean region (OECD, 2006) where irrigated agriculture is a major water user accounting for more than 60% of total resource. The concentration of irrigation in these countries is so noticeable that about 75% of the 16×10^6 ha of agricultural land equipped for irrigation in the EU concentrate in the Mediterranean countries France, Greece, Italy, Portugal and Spain (Eurostat, 2000, 2003). The high water demand of agriculture and population in the Mediterranean are exacerbated by the limited natural availability of water resources and high climatic variability (MGWWG, 2005; Wriedt *et al*., 2009). Looking at the origin of agricultural activity in these countries, some authors claim that two main drivers have affected water consumption, most acutely in the arid and semi-arid regions. First, a significant part of the construction cost of waterworks has been sustained by public funds; water in general and irrigation water in particular often necessitates large initial investments in infrastructure (Johansson *et al*., 2002; Molle & Berkoff, 2007a). Second, the Common Agricultural Policy (CAP) allowed farmers to purchase irrigation water at subsidised prices leading to contradictory effects in many rural areas (Berbel & Gutiérrez, 2004). On the one hand, the profits generated from irrigated land were significantly beneficial for farmers, yet the absence of incentives to sustainably use and conserve water resources led to negative externalities for the local aquatic system (Baldock *et al*., 2000). This high level of water need and abstraction has caused over exploitation of groundwater in several Mediterranean regions. The intensive development of groundwater resources has brought about significant social and economic benefits, but their unplanned nature has also resulted in negative environmental, legal and socio-economic consequences (Hernández-Mora *et al*., 2010). This situation is not exclusive to Spain. With different hues it is similar in most arid and semi-arid countries (Zoumides & Zachariadis, 2009). WWF/Adena (2006) estimate that 45% of all water abstracted from aquifers in Spain is abstracted illegally (and unrecorded). Reliable information is missing also in Greece (Panoras & Mavroudis, 1996; WWF, 2003) and Portugal (WWF, 2003).

Water quantity and quality connection

To understand water quality in SE countries it is necessary to understand its connection with water management. In these countries, and especially in Mediterranean basins, water quality is affected by many pressures, but the effect of all these is augmented by low flows in the rivers during the greater part of the year. Many rivers have low flows in winter because water is storage for the summer period when irrigation demands require high quantities of resource.

Low flows in rivers are due to the concentration of the demands in summer periods. These include irrigation of crops and tourism demand. Storage of water in headwater reservoirs during many months of the year is a practical issue in the management of arid and semi-arid basins to maintain the water supply reliability. The Mediterranean climate typically presents dry summers and wet winters with high temporal seasonal and inter-annual precipitation gradients. Furthermore, mean precipitation is notably less than in Central and Northern European areas. Droughts in SE last several years with devastating effects. As a consequence, problems of water scarcity (where demand exceeds the available resource) are widely reported. In 2007, the European Commission (EC, 2007) estimated that at least 17% of EU territory had been affected by water scarcity and put the cost of droughts in Europe over the previous 30 years at EUR 100 billion, with significant consequences for the associated aquatic ecosystems and dependent users (EEA, 2009, 2010a).

However, while EU legislation has extensively focused on water quality, there has been far less attention paid to the issue of water quantity. As a result, water quantity assessments are usually not based on data reported under EU legislation, and these assessments instead need to draw on a variety of different reporting (EEA, 2012b). A sustainable approach to water resource management, focusing on both water quality and quantity, requires that society conserves water and uses it more efficiently. Integral to this is a more equitable approach to water abstraction that addresses not only the requirements of competing economic sectors but also the requirements of healthy and resilient freshwater ecosystems (EEA, 2009, 2012a).

LLOBREGAT RIVER BASIN

As an example of a Mediterranean basin we present herein the water quality status of the Llobregat River basin (LRB). LRB is the most polluted basin in Spain and perhaps an extreme case of a problematic basin. Although it is not much affected by agriculture activities, urban and industrial supplies affect water quantity and quality of the water system. As depicted in Fig. 1, the LRB is located in Catalonia, Spain. It originates in the Pyrenean Mountains and flows into the Mediterranean Sea, south of the city of Barcelona. The LRB is 4948 km^2 in extent and has a population of over one million people. Its mean annual precipitation is 704 mm/year, which produces a mean flow of 694 hm^3/year in natural conditions. The main tributaries are the Cardener and Anoia rivers.

The main use of water is for human water supply to cities located in the basin, and to the influence area of Barcelona. The water supply to the influence area of Barcelona is performed mainly by the system of two rivers, namely the Ter and Llobregat rivers. This system supplies

Fig. 1 Location of study area and main elements.

water to approximately 5 million people. In the Llobregat River basin several important cities are located with important economic and industrial activities. Moreover, these cities discharge their wastewater into the Llobregat River, or one of its tributaries. There are more than 60 wastewater treatment plants (WWTP) in the basin. Industrial activity has a great influence on water quality outflow of the WWTP.

The second water use in the basin is supply to irrigation with an annual demand of 20 hm^3, which is mainly required between April and September. Irrigation demands are located in different areas such as: Anoia, Manresa and the lower part of the Llobregat. The main storage infrastructure is composed of three reservoirs: La Baells (115 hm^3), La Llosa del Cavall (115 hm^3) and Sant Ponç (24 hm^3). The former is located upstream on the Cardener River, and the other reservoirs are in the headwaters of the Llobregat River.

The critical points in the LRB are the two intakes of water supply for Barcelona: Abrera Treatment Plant (ATP), and the San Joan Despí Treatment Plant (SJTP). The ATP is located on the Llobregat downstream from the confluence with the Cardener. The SJTP is located in the lower part of the basin and has many water quality problems due to the effects of all the environmental pressures in the basin. Due to the low quality of the raw water, treatment in both plants is very expensive and the resulting quality of the treated water, although potable, is not very good.

Components and environmental pressures

An analysis of the water quality in the basin was performed using water quality data from the Agencia Catalana del Agua (ACA; Catalonian Water Agency; http://aca-web.gencat.cat/aca) monitoring network. This network has control points at several strategic points on the LRB, and it has monthly data available for 2002 to 2011. Moreover, data from several wastewater monitoring campaigns are available. Other available data include flow-gauging stations, consumption of water demands, and storage volumes in the reservoirs. In general, the water quality of the LRB is highly influenced by the WWTP effluents, mining activities and agricultural activities.

Following the schema of Fig. 2, upstream of the Llobregat River, the main pressuring activities are from agricultural demands and mining activities. Salt and potassium mining activities are the main cause of conductivity augmentation in the basin. At this point, the Llobregat River merges with the Cardener River, also affected by salt mining activities and by Manresa's WWTP. Downstream of its confluence with the Cardener River, two important point loads on the Llobregat River are from Castellbell i El Villar, and Monistrol. Currently, Monistrol waste is insufficiently treated, and it has a high discharge of organic matter. The ATP that takes water for the Barcelona supply is located downstream. This is one of the critical points in the basin for water quality. In this part of the river water quality is also affected by the discharges from the Abrera and Martorell WWTPs. Downstream from the city of Martorell, the Llobregat River merges with the Anoia River. The Anoia River is impaired from its headwater, where agricultural irrigation increases nitrates to high levels. Furthermore, the discharge from the Igualada WWTP (in Anoia River) impacts in all constituents because the load is approximately 60% of the flow in the river. In recent years, the water treatment at Igualada WWTP has improved and, consequently, so has the water quality of the river. However, Anoia River flows are currently diverted to a channel to avoid the water from the Anoia River decreasing the water quality downstream. In the lowest part, the SJDP intake for human supply is located, and it is at this point where water quality presents many problems for the human water supply and environmental uses. High concentrations of ammonia, nitrates and phosphates are common, and they require high levels of treatment to be applied to the water in order to supply the metropolitan area of Barcelona.

Current water quality situation

One of the main concerns for the whole system is the content of salt in the water. The main factors affecting conductivity are the natural geology, mining activities and industrial activities. Conductivity in the Llobregat and Cardener headwaters is approximately 500 μS/cm, but after the

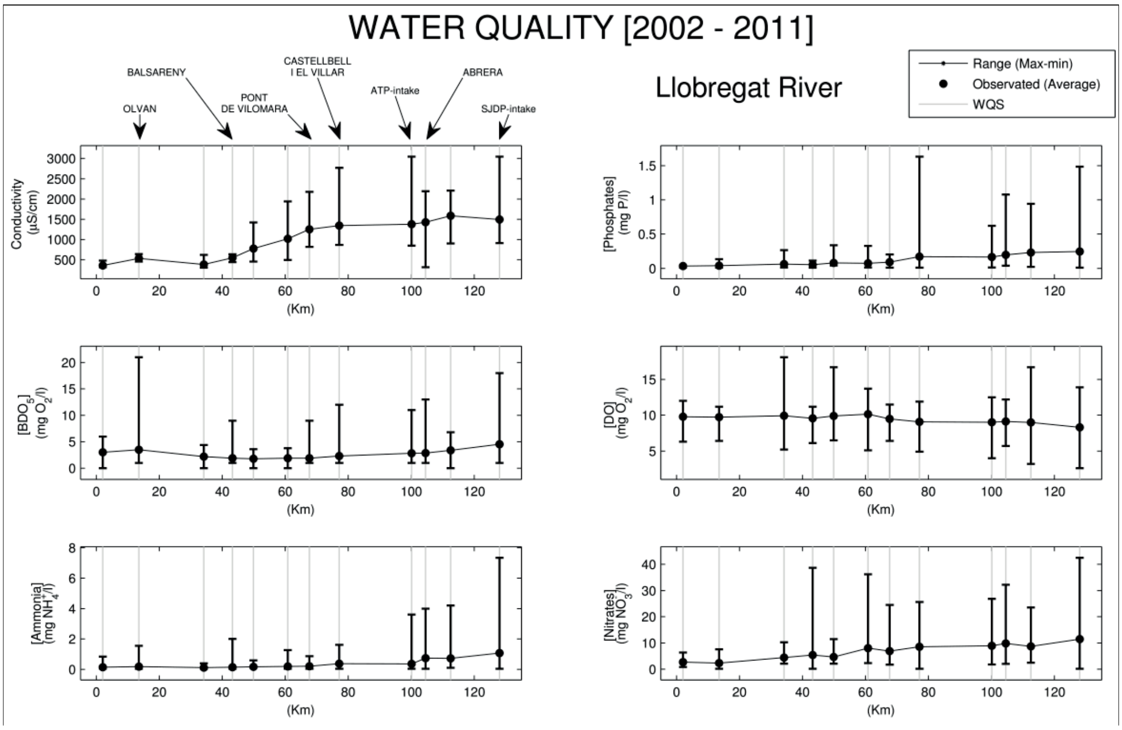

Fig. 2 Mean concentrations of several constituents measured in water quality stations of LLRS.

effects from mining activities, the measurements average 1200 µS/cm and have maximums of approx. 4000 µS/cm. This change is accompanied by high levels of conductivity for almost all of the WWTP loads in the basin, such as Navarclés and Sallent, with a range from 1000 to 11 000 µS/cm. As a consequence, conductivity is high in the entire WRS. At the intakes of the water supply, the average conductivities are 1360 and 1435 µS/cm for ATP and SJTP, respectively. The Anoia River has a similar behaviour, with high concentrations in the headwaters, and the values of conductivity at Jorba station range from 1200 to 3100 µS/cm. At the end of the Anoia River, conductivity averages 1930 µS/cm.

A sufficient supply of dissolved oxygen (DO) is vital for all higher aquatic life. In the middle and lower regions of the river, the addition of organic matter and ammonium concentrations slightly affects the DO levels. The most critical point in the basin with regards to DO concentrations in the Anoia River is located downstream of the Igualada effluent discharge where summer concentrations are usually near 4 mg/L, but in some years they are less than 2 mg/L. Another critical point could be downstream of the Monistrol WWTP effluent due to the amount of organic matter discharged; however, there are not enough data to corroborate this hypothesis.

In the LRB, concentrations of BOD have decreased in the last decade due to improvements in the treatment of wastewaters. The main WWTPs produce effluents with less than 20 mg/L of BOD_5. Measured concentrations of BOD_5 across the entire basin present values near or lower than 3 mg/L.

The average concentration plot (Fig. 2) demonstrates a continuous increase of nitrate concentrations in the basin ranging from values near 3 mg-NO_3^-/L to 10 mg-NO_3^-/L. These increases of nitrate are due to the effects of urban discharges and diffuse pollution from some agricultural areas. The maximum values of nitrate concentrations in the SJTP stations are approx. 22 mg-NO_3^-/L. In the Anoia River, it is remarkable that measures of nitrate concentrations at the headwater station, Jorba, are very high with values greater than 70 mg-NO_3^-/L, while downstream of the effluent from the Igualada WWTP, paradoxically, nitrate concentrations drop, and finally, groundwater inflows into the medium-low part of the river result in an increase of the nitrate concentrations again.

Phosphorus enrichment is also a general problem in the basin. WWTP effluents are the main source of phosphorous. The average values of the phosphates along the river range from 0.01 to 0.25 mg-P/L. Maximum values at the ATP and SJTP stations are close to 1.5 mg-P/L. In the Cardener, the main source of phosphorous is the load from the Manresa WWTP, and it increases phosphate concentrations to over 0.2 mg-P/L with maximums of 1.2 mg-P/L. In a similar way as in the Anoia River, the Igualada WWTP is the main source of phosphorous, but in this case, concentrations in the river reach 8 mg-P/L.

One of the main water quality problems in the LRB is the ammonia concentration. Concentrations of over 1 mg/L commonly appear at several points in the basin, mainly due to the effect of urban loads. This is the threshold established by the ACA that distinguishes a good or bad water quality status for the water body. In recent years, different nitrogen release treatments have been incorporated into some WWTPs. The treatment efficacy has been reflected in several areas of the basin where ammonia concentrations have decreased. For example, at the Castegallí station on the Cardener River, the ammonia concentrations were reduced in recent years from 1–5 mg/L to values lower than the threshold. A similar situation has occurred at the Vilanova del Camí station in the Anoia River. However, some of the nitrogen treatments developed did not work well during a transitional period, especially in winter, due to low temperatures. For example, the concentrations of ammonia in the loads from the Abrera WWTP were 4.9 mg-NH_4^+/L in August and 30 mg-NH_4^+/L in December. In the middle and lower region of the LRB, the concentrations of ammonia are high, and they increase from upstream to downstream. In the ATP and SJTP water quality stations, the concentrations are often over 1 mg NH_4^+/L with maximums of 3 and 5 mg-NH_4^+/L, respectively, in drought periods.

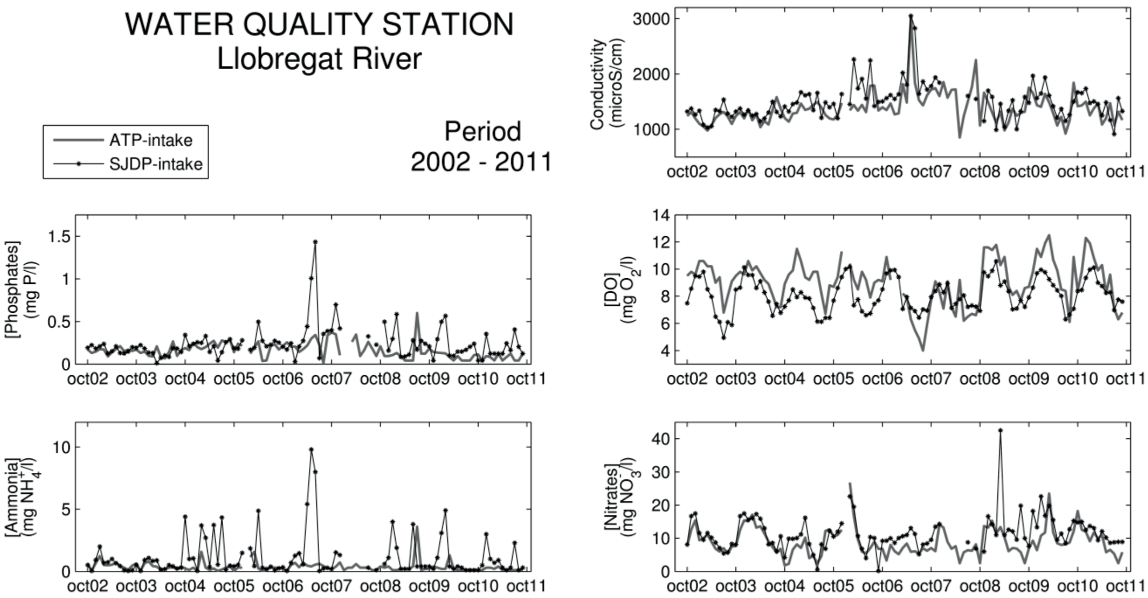

Fig. 3 Concentrations of several constituents in ATP and SJDP water quality stations.

However, the water quantity problems are equal, or even more important than water quality ones. The LRB suffers from recurrent drought periods that affect the human water supply for Barcelona and the population of the Llobregat basin. The last drought period, 2005–2008 had high social, political and economic consequences, and tough measures were adopted to mitigate drought consequences. It was necessary to carry water by ship from a desalination plant located 350 km away. In the drought period, releases from the reservoirs were reduced to the minimum and tributaries flows were null. In this situation, the dilution of the discharges and self-purification of the river were minimized. Consequently, peaks of ammonia concentrations, and other contaminants, occurred at the water intake locations, complicating water supply treatment.

LRB water quality presents a serious problem during drought periods when storing water in reservoirs reduces river flows, improving the reliability of water supply to Barcelona city whilst decreasing water quality and the environmental condition of many water bodies in the basins.

CONCLUSIONS

This paper summarizes water quality problems of Southern Europe countries and their main drivers. We have tried to remark on several points:

- Although these countries present similar features that determine water quality, the essential regional different conditions are important to consider.
- Lack of data is a common problem in extracting conclusions from European and database reports.
- Historical water treatment investments have been different among Mediterranean countries affecting the current status of the water bodies.
- River flows are a major factor, and as important as water treatment, in the status of the water quality of the water bodies. The link between water quality and quantity takes on special importance in these basins.
- Agriculture, and especially irrigated areas, in Southern Europe is a twofold problem for water quality. First, as a pollution source producing nitrates, pesticides and other contaminants. Second, because the large amount of water necessary for this activity affects river flows and water quality during several months of each year.

As an example, we described current water quality status of Llobregat River Basin (Spain), where water quantity and quality are serious problems for environment and human activities.

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