REPORT



Potential uses of pumped urban groundwater: a case study in Sant Adrià del Besòs (Spain)

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Abstract Urban groundwater has often been over-exploited for industrial uses. Now, this usage tends to be reduced or the resource abandoned due to pollution and/or changes in land use. The use and the subsequent disuse of groundwater has resulted in rising water tables that damage underground structures (e.g., building basements and underground car parks and tunnels), leading to the need for additional pumping in urban areas. In the case of the underground parking lot of Sant Adrià del Besòs (Barcelona, NE Spain), large amounts of urban groundwater are pumped to avoid seepage problems. Can this pumped groundwater be used for other purposes (e.g., drinking water and urban irrigation) instead of wasting this valuable resource? To answer this question, it was necessary to quantify the groundwater recharge and to assess the evolution of groundwater quality. The limiting factor at this study site is the groundwater quality because ammonium and some metals (iron and manganese) are present at high concentrations. Hence, further treatment would be needed to meet drinking water requirements. The pumped groundwater could also be used for supplementing river flow for ecological benefit and/ or for mitigating seawater intrusion problems. Currently, only

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² GHS, Institute of Environmental Assessment & Water Research (IDAEA), CSIC, Barcelona, Spain a small amount of this urban groundwater is used for cleaning public areas and watering public gardens. This situation highlighted the urgent need to manage this resource in a responsible and more efficient manner, especially in moments of high water demand such as drought periods.

Keywords Drinking water resource · Groundwater management · Urban groundwater · Water quality · Spain

Introduction

Rapid urbanization coupled with industrial activities has increased the demand for water resources in the last few decades (Schirmer et al. 2013). This situation is even more evident in large cities, where half of the world's population currently lives, and where an increase of 2.5 billion urban dwellers is expected by 2050 (UN 2013). Urban groundwater, with increasing frequency, is considered to be a valuable resource for the water supply of cities and a key factor in the management of urban areas (Howard and Israfilov 2002; Drangert and Cronin 2004; Foster and Hirata 2012), especially during drought periods in semi-arid regions such as the Mediterranean coast of Spain.

Drought is relatively common in this region and can lead to negative consequences—for example, water for supply purposes had to be transported from a far river by sea tankers and some water uses were banned by the authorities during the last severe drought that occurred in Barcelona in 2007–2008, after a dry period of 42 months (Martin-Ortega and Markandya 2009; March et al. 2013; Parés et al. 2013). These measures obviously affected the citizens and the economy. The possibility to take advantage of urban groundwater is only seriously considered when other water sources (e.g., surface water) are

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scarce. Generally, when drought periods end, urban groundwater is forgotten and it is not considered within water management schemes. In fact, urban groundwater derived from temporal and continuous pumping is generally discharged into sewers, which implies a loss of resource and energy, and can lead to deterioration of the sewer systems.

The aquifers of Barcelona (in north-eastern Spain) have been seriously affected by the city development. Since the early 20th century, the aquifers underneath Barcelona were subjected to heavy water extraction for industrial purposes and this produced groundwater-level drawdowns of up to 15 m (Vázquez-Suñé et al. 2005). This fact was not considered in the construction of underground infrastructures and some of them were constructed below the natural piezometric level. The aquifer overexploitation also led to groundwater quality problems due to seawater intrusion (Vázquez-Suñé and Sánchez-Vila 1999). In the 1970s, groundwater extraction (for industrial and drinking purposes) decreased due to the pollution problems and/or changes in land uses (i.e., relocation of the industries), and groundwater levels progressively rose as a result (Vázquez-Suñé et al. 2005). The continuous recovery of groundwater levels has posed a serious threat to

some underground urban structures (e.g., damage and flooding problems) that were designed and constructed when minimum groundwater levels occurred (Custodio 2001; Vázquez-Suñé et al. 1997). A common procedure for controlling these problems is to pump large volumes of groundwater (Pujades et al. 2014; Wong 2001).

This is the case of the three-story underground parking facility located in the plaça de la Vila at Sant Adrià del Besòs in Barcelona (Fig. 1). This parking lot was constructed during the overexploitation period and it was opened in 1971. A few months later, the parking lot had several seepage problems and in December of the same year, a single flood event covered one and a half stories after a period of heavy rainstorms. The parking lot continuously had seepage problems thereafter, and thus, pumps were installed pumping at a flow rate of about 250-300 L/s (Ondiviela et al. 2005). This situation was not sustainable due to the high economic costs that included the maintenance of the pumping equipment, and the City Council of Sant Adrià del Besòs decided to remodel the parking area to minimize the groundwater abstraction. A new two-story car park was constructed in 2005, and a more efficient drainage system was designed. The current drainage



Fig. 1 a Location of the study area and b spatial distribution of the observation points (section A-A'). c Schematic description of the hydrogeological conceptual model as well as the screen depths of the

pumping well and the observation points. d The piezometric surface from the River Besòs to the parking area. Note that the piezometric level is in meters above the sea level (masl). (Figure modified from Jurado et al. 2015)

system pumps approximately 150 L/s and most of this groundwater is directly discharge into the sewage system. Although the new drainage system is much better than the initial one, a large volume of water is still wasted. In addition, the continuous pumping implies an economic cost because of the continuous energy consumption and aspects related to the waste-water operation such as the maintenance of the sewer system, the increase of the pumping costs and the dilution of water in treatment plants. It is clear that there is high availability of groundwater resources in Sant Adrià del Besòs aquifers, but these resources are currently not efficiently managed. This groundwater could be used for covering various aspects of the growing demand (i.e., droughts periods), becoming a strategic water supply resource. This last consideration requires the evaluation of groundwater recharge and the groundwater quality over time to anticipate the evolution of the pumped urban groundwater facilitating the decision making and improving its management.

This report presents a set of tools to assess the potential uses of urban groundwater by means of numerical and mixing models. To reach this objective, it was necessary to (1) quantify the groundwater recharge, (2) characterize the sources of contamination, (3) investigate the temporal evolution of groundwater quality in relation to the sources, and (4) assess the potential uses of the urban groundwater. The approach was applied to the aquifer of the Besòs River Delta but it can be applied to other urban aquifers with similar purposes.

Materials and methods

Site description

The Besòs River Delta, with an area of approximately 20 km^2 , is located near Barcelona in the north east of Spain. The delta has an average slope of 1% and 12 km of coastline, bordering the Mediterranean Sea in the south east (Fig. 1). The study area corresponds to the lower part of the delta at Sant Adrià del Besòs city (Fig. 1) and it is a densely populated area.

Previous works carried out on the aquifers of the low part of the Besòs River Delta

Vázquez-Suñé et al. (2010) identified and quantified the recharge sources in the aquifers of Barcelona, including the Besòs River Delta area, by means of end-member mixing analysis (EMMA). They identified five different recharge sources in the Besòs River Delta area using hydrochemical data (i.e., chloride, sulphate, $\delta^{18}O_{H2O}$, δD_{H2O} , $\delta^{34}S_{SO4}$, boron, fluoride, bromide, zinc, total nitrogen, residual alkalinity and EDTA). These recharge sources include inflows from surfacewater bodies, seawater intrusion and water from the heavily polluted River Besòs. There are also three groundwater recharge sources from anthropogenic activities: (1) loss from the water supply network, (2) loss from the sewer system and (3) the runoff water in paved areas that recharges the aquifers through sewer seepage and/or direct infiltration. Finally, direct rainfall recharge occurs in the non-urbanized areas in the mountain range. Vázquez-Suñé et al. (2010) quantified that 42% of the average groundwater recharge in the delta was from the river followed by loss in the sewage network (28%) and rainfall recharge in the non-urbanized area (17%). Minor contributions to groundwater recharge were loss in the water supply network (9%) and runoff water in paved areas (4%). The results of the study were based on a single sampling campaign and, therefore, groundwater composition did not change significantly; however, Vázquez-Suñé et al. (2010) pointed out that variations in groundwater composition could be expected in areas where sources of recharge can change significantly in volume and composition. This is the case of the River Besòs, which is characterized to have irregular flow patterns reflecting seasonal changes in river-water quality.

Tubau et al. (2014) carried out an EMMA to identify the minimum number of river end-members needed to explain the temporal variability of the River Besòs using a large hydrochemical dataset from the Catalan Water Agency (ACA 2016) at the Santa Coloma gauging station (Fig. 1). Tracers used were chloride, sulphate, sodium, bicarbonate, calcium, magnesium, potassium, nitrate, ammonium, total nitrogen and electrical conductivity. Three river end-members were selected: two corresponding to the dry season (D1 and D2) and one to the wet season (W1). The wet end-member (W1) is related to short but intense rainfall events, and the dry end-members are related to the null or low levels of rainfall that occur during the rest of the year, especially in summer (Tubau et al. 2014). The wet river end-member (W1) presented lower concentrations than those of the dry periods (D1 and D2) for all species except for dissolved oxygen, ammonium and nitrate. Two dry river end-members were necessary to explain the variability of the nitrogen species in dry periods-for example, the concentrations of ammonium are 31.5 mg/L for D1 and 3.6 mg/L for D2. The composition of these end-members is summarized in Table S1 of the electronic supplementary material (ESM). Afterwards, these river endmembers were used for quantifying the groundwater recharge at the Besòs River Delta aquifer in Sant Adrià del Besòs village (Jurado et al. 2015; Tubau et al. 2014). These studies concluded that dry season end-members dominated over the wet season end-member, in a proportion 4:1. Similarly, Jurado et al. (2013) quantified the groundwater recharge at the same area by means of mixing analysis but considering the following recharge sources: river (D and W end-members), loss from sewage network and urban runoff. Tracers used were chloride, sulphate, total nitrogen, $\delta^{18}O_{H2O}$, δD_{H2O} , $\delta^{34}S_{SO4}$, $\delta^{18}O_{SO4}$, $\delta^{13}C_{DIC}$, electrical conductivity and bicarbonate. River Besòs was by far the largest contributor to the total recharge,

representing 88% in total (60% from dry conditions and 28% from wet conditions), whilst network sewage loss and runoff water in paved areas represented 6% each. In the same area several processes were quantified in the river–groundwater interface using mixing analysis (Jurado et al. 2015). The processes that occurred when river water infiltrated the aquifer were redox processes (e.g., aerobic respiration and denitrification) and carbonates dissolution. Finally, other studies developed in this aquifer were concerned with the occurrence of organic micropollutants such as surfactants, pharmaceutical compounds, drugs of abuse and UV filters (Jurado et al. 2012, 2014a, b; López-Serna et al. 2013; Serra-Roig et al. 2016; Tubau et al. 2010).

In the present work, the previous works carried out in Besòs River Delta aquifers were extended in two aspects. Firstly, this study quantifies the groundwater recharge from the river to the parking area at Sant Adrià del Besòs (section A–A', Fig. 1). Secondly, none of the previous works assessed the groundwater quality over the time and proposed alternative uses for the pumped urban groundwater; the present study addresses these shortfalls.

Hydrogeological setting

The fluvio-deltaic sediments of the Besòs River Delta lay over a basement composed mainly of slates and granites from the Palaeozoic and clays from the Pliocene (Velasco et al. 2012). These sediments make up two superposed aquifer layers: (1) the shallow aquifer, which is an unconfined aquifer made up of siliceous sands and carbonate gravels, and (2) the main aquifer, which is a confined aquifer formed of siliceous and carbonate sands. These aquifers are separated by an aquitard made up of clays and silts.

The shallow aquifer is hydraulically connected to the River Besòs at Sant Adrià del Besòs village (Fig. 1). The River Besòs presents a Mediterranean hydrological pattern with an irregular and torrential regime and, in many cases, extremely flashy floods. Average streamflow is 4 m³/s at the Santa Coloma gauging station (Fig. 1), and the maximum flow occurs in the form of floods with discharges reaching up to 100 m^3 /s. These floods vary within the year and from 1 year to another, but they mostly take place in autumn and spring. In contrast, the flow rate in summer is mainly due to the effluents of waste water treatment plants (WWTPs), thus worsening the water quality. Overall, river-water quality is better after rain events than in dry periods. Figure 2a shows the flow versus the chloride concentration of the River Besòs at the Santa Coloma gauging station (Fig. 1). Chloride fluctuations depend on the rain, being low after flood events (e.g., May 2008, February 2009 and May 2010, among others). Besides, the river presented higher chloride concentrations during the drought period (July 2007-April 2008) than those from May 2008 to July 2014 (average concentrations were 266 vs. 198 mg/L).

The aquifer-river system is very dynamic and it is enhanced by the short residence times that range from 3 weeks to 1 month from the river to the underground car park (Jurado et al. 2015). Groundwater flows from the River Besòs to the parking area because the current drainage system pumps 150 L/s to avoid seepage problems (Fig. 1). The drainage system is composed of four pumping wells (ADPM, ADPQ, ADPR and ADPW, Fig. 1) constructed in the surroundings of the parking area. There is also a monitoring network of 16 observation points that, together with the pumping wells, are used for assessing groundwater head variations and the quality of the groundwater. The six sampling points considered in this study are the well ADPW and the observation points from the series SAP (located close to the river) and ADS-n (Fig. 1). The screen depth intervals for the observation points are 9.5-11.5 m for SAP-1 and SAP-2, 4.5-6.5 m for SAP-2b and 3-15 m for ADS-6n and ADS-7. The aforementioned irregular flow pattern of the river is reflected in the piezometer hydrographs (Fig. 2b)-for example, there were heavy rain events in March 2011 that caused an increase in groundwater levels of 0.36 and 0.28 cm at observation points SAP-1 and ADS-8, respectively.

Sampling and analytical methods

A total of six field campaigns were carried out between July 2007 and July 2014 (July 2007, February 2008, October 2008, May 2010, December 2013 and July 2014; Fig. 2). Thirty-five samples were collected from groundwater (SAP-1, SAP-2, SAP-2b, ADS-6n, ADS-7 and ADPW), and six were from the River Besòs. Before sampling, wells and observation points were purged by pumping three well volumes to remove the stagnant water. Temperature, electrical conductivity, pH, Eh, and dissolved oxygen were measured within a flow cell after reaching stability (EPA Victoria 2000). Samples were collected after stabilisation of field parameters, stored in a field refrigerator and taken to the laboratory at the end of the day.

Samples were analysed in two different laboratories, and they were treated or not in the field according to the requirements of each laboratory. The samples from the first to the fourth sampling campaigns (C1–C4) were analysed at the laboratory of the ATLL (Aigües Ter-Llobregat) in Barcelona. In this case, the samples were filtered and acidified in the laboratory facilities for the analysis of cations. Cations were analysed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS), and the solution was acidified with 1% (v/v) HNO₃ and centrifuged to 3,500 rpm prior to the analysis. Anions were analysed using ion chromatography (IC). Bicarbonate was analysed manually by chemical evaluation with sulphuric acid accounting for the pH of the sample. Ammonium was analysed by spectrophotometry based on the indophenol blue method. Total organic carbon (TOC) was analysed using the **Fig. 2** a River Besòs flow (m³/s) versus chloride concentrations of the river from July 2007 to July 2014. **b** Flow (m³/s) versus groundwater level (masl) at observation points *SAP-1*, *ADS-6n* and *ADS-8* from September 2005 to September 2014. Chloride concentrations and river flow were measured by Catalan Water Agency (ACA) at the Santa Coloma gauging station (Fig. 1)



680 °C combustion-infrared method with a platinum catalyst and using a non-dispersive infra-red (NDIR) detector. The analysis of pesticides and chlorinated solvents were carried out using gas ion chromatography (GC) with an electron capture detector (ECD). The samples from the last two campaigns (C5 and C6) were analysed at the IDAEA-CSIC laboratory facilities. In these campaigns, samples were filtered and acidified in the field when required (for cation-trace analysis, samples were acidified with 1 ml of 20% diluted nitric acid for sample preservation), and total alkalinity was determined in the field by acid-base titration using the Aquamerck Alkalinity Test. Anions were analysed by high performance liquid chromatography (HPLC), and cations were analysed by coupled plasma atomic emission spectrometry (ICP-AES). TOC samples were collected in glass bottles previously muffled and filtered through a 0.45-µm nylon filter and acidified with 1 ml of HCl (2 N); the bottles were sealed with Parafilm® to minimize any contact with air. TOC was analysed by the catalytic oxidation method at 680 °C using a Shimadzu TOC- V CSH instrument. Ammonium was analysed with a selective electrode (Orion 9512). The limits of detection for water samples set up by both laboratories (ATLL and IDAEA) for minor and major ions, metals and organic compounds (pesticides and chlorinated solvents) are summarized in Table S2 of the ESM.

Quantification of the groundwater recharge and residence time

A numerical groundwater flow model was built using the finite element code TRANSIN-IV (Medina and Carrera 1996, 2003; Medina et al. 2000) with the visual interface VISUAL TRANSIN (UPC 2003) to evaluate the river infiltration rate into the aquifer and to compute the groundwater residence time (from the river to the parking area; section A–A', Fig. 1).

The numerical model is a cross plot section that spans from the river to the parking area and it simulates the groundwater flow in the shallow aquifer of the Besòs River Delta (section

A-A', Fig. 1). Groundwater flow in/from other directions was not modelled because pumped groundwater quality mostly depends on the river water. The problem was simulated with a two-dimensional (2D) numerical model in which a partially axisymmetric condition was implemented. It is assumed that the pumping in the parking area induced a drainage divide where: (1) the groundwater flow around the parking is practically radial and only linear near the river, and (2), approximately half of the pumped groundwater infiltrates the aquifer in the river section located in front of the parking. Consequently, the axisymmetric condition was implemented by multiplying the flow and transport parameters by πr , where r is the distance from the parking. Dirichlet boundary conditions (BCs) were adopted in the parking area and in the riverbed (Fig. 3), where a detailed registers of the piezometric head from the observation point ADS-8 and of the River Besòs levels were available. A no-flow BC was assumed on the bottom of the numerical model because materials underlying the modelled domain have low hydraulic conductivity; thus, vertical flow through them is negligible.

The thickness of the modelled domain ranged from 7 m in the middle to 12 m in the river boundary. The shallow aquifer was divided in four layers (L1-L4) according to their hydraulic conductivities (Fig. 3). L1 represents the riverbed sediments and is made up by silts while the other three layers are mainly constituted by sands and/or gravels. Flow and transport parameters were obtained from De Buen (2009). Hydraulic conductivities of layers L1-L4 were 0.75, 768, 87 and 348 m/day, respectively, and the storage coefficient was 0.14 in the whole aquifer. Regarding the transport parameters, 0.1 and 0.01 m were used, respectively, as longitudinal and transversal dispersivities for all layers, and porosity was 0.21 in the whole domain. The observation points were implemented in the numerical model as an area of high hydraulic conductivity located at the depth of their respective screens. The mesh of the numerical model consisted of 3,012 nodes and 5,540 triangular elements whose vertical and horizontal size was 1 m over the whole domain. The period simulated was from September 2005 to September 2013 in time steps of 0.5 days.



Fig. 3 Boundary conditions (BCs) and layers (L1-L4) of the numerical model

Assessment of the evolution of groundwater quality

The MIX code (Carrera et al. 2004) was used to assess the evolution of groundwater quality. The methodology consists of the following steps: (1) the identification of the potential recharge sources and the selection of the appropriate tracers and (2) the evaluation of the mixing ratios in groundwater over the time.

MIX code computes mixing ratios in cases of uncertain end-members using the concentration of mixed samples to reduce the uncertainty. Uncertainty is related to sampling and analytical errors, lack of data to characterize the source composition and/or the occurrence of geochemical processes. This uncertainty is quantified through covariance matrices, which requires defining the reliability of the measurements and the results depend on the assumed standard deviations. In fact, standard deviations must be carefully assigned in both observation points and recharge sources. Generally, standard deviations are selected depending on whether or not the tracers are conservative-for instance, higher standard deviations are assigned to the non-conservative species (e.g., nitrate) rather than those assigned to the conservative species (e.g., chloride) because the former can participate in geochemical processes.

The algorithm to estimate mixing ratios follows the next steps: (1) definition of initial mixing ratios by conventional least squares assuming that the concentrations of endmembers are known, (2) given these mixing ratios, maximise the likelihood function to estimate the expected values of mixture and end-member concentrations; (3) given the expected values of mixture and end-member concentrations, maximise the likelihood to obtain the mixing ratios and (4) repeat steps 2 and 3 until convergence. Further details on the description of these steps and the equations involved in this algorithm can be found in Carrera et al. (2004).

Results and discussion

Quantification of the groundwater recharge and the residence time

The results of the model are the quantification of the groundwater recharge and the evaluation of the residence time. On the one hand, the average recharge from the river is 95.4 L/s in the modelled profile. This means that 56% of the pumped water was recharged from the section of the river located near the parking area from September 2005 to September 2013. Consequently, an average volume of 3×10^6 m³/year of urban groundwater was directly poured into the sewage system instead of being used more adequately (Fig. 4). This volume varied from 1 year to another, with a maximum of 4.7×10^6 m³ in 2006 and a minimum of 2.3×10^6 m³ in 2008.





These volumes can be directly related to the pumping carried out in the parking area-for example, the demolition of the old parking area started in February 2006, and thus, additional pumping was required until February 2007 in order to construct the actual parking lot. Conversely, the minimum volume recharged was during the aforementioned drought period. Computed versus measured head plots of the observation points SAP-1, ADS-6n and ADS-7 are shown in Figure S1 of the ESM. On the other hand, the minimum residence time—i.e., with a retardation coefficient (R) of 1—was computed by simulating an instantaneous injection of a conservative tracer in the river boundary and observing the breakthrough curve in the parking boundary (Fig. S2 of the ESM). Results show that the concentration peak (recharged in the river boundary) reaches the pumping area after 20 days. Ferrer (2006) and De Buen (2009) estimated R values of 2.5 and 5.1, respectively, for the shallow aquifer. Considering these R values, the residence times would range from 50 to 100 days to cover the distance between the river and the parking area (section A-A', Fig. 1).

The results of the numerical model are of paramount importance for the urban groundwater management in the study site. Firstly, a large volume of the pumped water comes from the section of the river located just in front of the parking lot (section A–A', Fig. 1); thus, the pumped groundwater quality depends on the river conditions. This fact would allow predicting changes in the pumped groundwater quality by monitoring the river quality. River-water samples are collected on a monthly basis by the ACA. Secondly, computed residence times provide information about the time for which quality changes observed in the river would be observed in the pumped groundwater. The minimum and maximum times needed by a compound infiltrated through the river boundary to reach the pumping wells range from 20 to 100 days, respectively. For instance, if a pollution episode occurs in the river, the use of groundwater should be stopped (or changed) between 20 and 100 days after this pollution event (note that for safety reasons, only the maximum retardation time is considered). In the same manner, better groundwater quality periods, which occur after wet periods (i.e., rain events), can be identified.

Assessment of the groundwater quality

Hydrochemistry of the River Besòs

River-water temperatures ranged from 11.5 to 28 °C, with an average value of 19.5 °C. Electrical conductivity and pH values varied from 914 to 1805 μ S/cm and from 7.8 to 8.7, respectively. River water presented high concentrations of dissolved oxygen (average was 7.8 mg/L) and total organic carbon (ranging from 6.6 mg/L in C4 to 11.5 mg/L at C2).

Major ion compositions showed that river water was of Cl-Na-(Ca)-(HCO₃) type accounting for all sampling campaigns except for C4, which was of HCO₃-Ca-(Na) type. The average concentrations of the River Besòs for all sampling campaigns are summarized in Table S3 of the ESM. Overall, riverwater composition was more mineralized in C1, C2 and C3 than in the last three campaigns due to the severe drought period that occurred in 2007-2008. This fact was reflected in the concentrations of chloride, sulphate, bicarbonate, calcium, magnesium, sodium, potassium and ammonium concentrations which were significantly higher than the last campaigns (C4-C6). In contrast, low concentrations were found in C4, C5 and C6 because they were collected during (C4) or just after (C5 and C6) rain events that caused a dilution effect (Fig. 2a). The lowest concentrations were found in C4 for most of the tracers, being the most significant differences with respect to the highest concentrations (C1 and C2) for ammonium (2 vs. 22.6 mg/L), chloride (96.9 vs. 323.2 mg/L), sodium (70.8 vs. 255.9 mg/L) and potassium (11.4 vs. 31.7 mg/L) (Table S3a of the ESM).

Concerning metals concentrations, the highest concentrations were for boron and iron (both reaching values around 0.25 mg/L) followed by manganese and aluminium, especially in the sampling campaigns C1 and C2. The concentrations of these metals were lower in the last three sampling campaigns. The rest of the metals were detected at low (maximal concentration was 0.046 mg/L for nickel at C2) or even null concentrations (e.g., cadmium from C1 to C4). The average concentrations (considering all the river samples) were lower than 0.01 mg/L for arsenic, cadmium, chromium, copper and lead (Table S3b of the ESM).

Regarding the organic compounds, a total of 26 pesticides were analyzed from C1 to C4 (pesticides included are listed in the ESM). Among them, 21 were not detected in any sample and the highest average concentrations were for diazinon $(4.4 \times 10^{-5} \text{ mg/L})$ followed by terbuthylazine $(2.6 \times 10^{-5} \text{ mg/L}, \text{TBA})$ (Table S3c of the ESM). The chlorinated solvents tetrachloroethene (PCE) and trichloroethene (TCE) were also detected in the river at average concentrations of 7.3×10^{-4} and 4.4×10^{-4} mg/L, respectively.

Hydrochemistry of the groundwater

Groundwater temperatures varied from 14.5 to 24.1 °C, with an average value of 19.8 °C, and pH values ranged from 6.9 to 7.6 (average was 7.2). Conversely, Eh values presented high variation, ranging from -5 to 371. Electrical conductivity values were between 1,277 and 1,780 µS/cm.

The range and the average concentrations for each sampling campaign (C1–C6) for the chemical compounds analysed are summarized in Tables 1, 2 and 3. Concentrations of these compounds are compared with the Spanish regulations for drinking water (RD 140/2003). Major ion compositions showed that groundwater in the shallow aquifer of the Besòs River Delta was of the Cl–(HCO₃)–Na–(Ca) type. The average concentrations for most of the major ions followed the same trend as the river concentrations, presenting little variation between field campaigns C1, C2 and C3. In contrast, these ions had, on average,

lower concentrations in the three last sampling campaigns (C4–C6). The most significant differences were for sodium, potassium and chloride. When considering the average concentrations of the major ions in all of the sampling campaigns (Cm, Table 1), none of them exceeded the Spanish drinking water guidelines (RD 140/2003); however, approximately 35% of the sampled points had chloride and sodium concentrations that were above this limit in sampling campaigns C1, C2 and C3.

Concerning nitrogen species, average nitrate concentrations ranged from 0.8 mg/L (C4) to 7.3 (C6) mg/L, and ammonium concentrations ranged from 2.2 mg/L (C6) to 8 mg/L (C1). It is important to highlight the moderate increase in the average concentration of nitrate in C5 and C6 (5.9 and 7.3 mg/L, respectively). Conversely, the average ammonium concentrations decreased in these sampling campaigns (2.7 and 2.2 mg/L, respectively). None of the sampling points exceeded the threshold of 50 mg/L of nitrate set by the RD 140/2003 for drinking water; however, all of the sampled points presented ammonium concentrations that were above the Spanish limit for drinking water (0.5 mg/L).

Regarding the average concentrations of metals, some of them such as iron, manganese and boron were detected at high concentrations of 0.42, 0.26 and 0.22 mg/L, respectively (Table 2). Arsenic was also detected at significant

Table 1Concentration ranges and average concentrations (*in parentheses*) of major ions, ammonium, dissolved oxygen and total organic carbon foreach sampling campaign (C1–C6) in mg/L, and electrical conductivity (μ S/cm)

Chemical parameter	Sampling campaign							RD 140/
	C1	C2	C3	C4	C5	C6		2003
Chloride	241.5–308.3 (268.1)	242.3–297.6 (268.8)	249.7–278 (259.7)	181.7–211.8 (199.5)	171.9–221 (191.8)	178.9–213.4 (192.9)	229	250
Sulphate	151.7–172 (164.4)	177.3–181.4 (179.3)	155.3–166.5 (160.7)	139.1–165.1 (150.3)	135.2–158.8 (146.8)	122.7–142.1 (135)	155.8	250
Bicarbonate	401–455.9 (428.1)	425.4-470.5	403.5-442.5	391.3–433.9 (412)	397.8–418.1 (406)	367–410 (399.6)	419.1	-
Calcium	131.4–146.8 (138.6)	137.5–149 (142.7)	129.4–147.9 (138.7)	117.3–135.4	106.4–120.7 (114.6)	92.3–120.9 (111.4)	128.7	-
Magnesium	28.2–29.6	23.6–33	28.1–31.9 (29.6)	25.2–28.7	21.2–25.3 (23.5)	19.8–25.4	26.9	-
Sodium	204–257.3	188.2-211.4 (200)	197.4–224	146.3–165.4	(119.5 - 153.4) (137.3)	120.5-157 (133.8)	175.6	200
Potassium	16.3-28.1	18.2-21.2 (20.1)	16.4-24	14.9-21.3	10.5-15.7 (13.4)	11-21.2 (14.5)	17.9	-
Nitrate	0-14.3 (2.9)	(2011) 0-24.7 (4.3)	<loq-10.1 (2.3)</loq-10.1 	0-3.7 (0.8)	0-12.4 (5.9)	0-13.2 (7.3)	3.9	50
Ammonium	2–15.2	4.7–11.8	4-9	2.1–6.3	1.2-5.8 (2.7)	0.6–6.6	5.1	0.5
Electrical conductivity	1,501-1,780 (1.631.2)	1,581-1,757 (1.654.8)	1,552–1,646	1,277-1,438 (1.375.5)	1,293-1,460 (1.360.2)	(1,389-1,574)	1,506.9	2,500
Total organic carbon	2.6–6.1 (3.9)	3-6.6 (4.3)	2.2–4.7 (3.4)	2.5–4.5	1.5–3.4 (2.3)	1.5–5.2 (2.9)	3.3	-
Dissolved oxygen	0.03–0.24 (0.01)	0.04–1.1 (0.4)	0.1–0.2 (0.1)	0.65–2.3 (1.2)	0.1–1.4 (0.3)	0.15–0.19 (0.2)	0.4	_

Note that *Cm* is the average concentration considering all of the sampling campaigns. *RD 140/2003* represents the recommended limits for Spanish drinking water guidelines. *LOQ* limit of quantification

 Table 2
 Concentration ranges and average concentrations (*in parentheses*) of metals for each sampling campaign (C1–C6) in µg/L

Metal	Sampling campaign							RD 140/2003
	C1	C2	C3	C4	C5	C6		
Aluminium	7.1–287.6	0.9–601.3 (161.6)	0–59.26 (17.6)	0–≤LOQ	5.5–23.5 (13.8)	9.5–67.1 (38.4)	53	200
Arsenic	2.4–31.1 (14.8)	4–27.5 (17.1)	3.6–32.2 (16.6)	2.1–17.3 (10.8)	1.6–9.1 (5)	1.5–14.5 (6.4)	11.7	10
Boron	260.7–283.3 (270.2)	229.2–298.9 (266.8)	199.3–264.9 (231.4)	138.2–201.7 (177.6)	163.4–244.3 (207.9)	171.6–223.5 (202.4)	224.8	1,000
Cadmium	<loq< td=""><td><loq< td=""><td>0</td><td>0</td><td><loq< td=""><td><loq< td=""><td>0.4</td><td>5</td></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td>0</td><td>0</td><td><loq< td=""><td><loq< td=""><td>0.4</td><td>5</td></loq<></td></loq<></td></loq<>	0	0	<loq< td=""><td><loq< td=""><td>0.4</td><td>5</td></loq<></td></loq<>	<loq< td=""><td>0.4</td><td>5</td></loq<>	0.4	5
Chromium	7.2–12.8 (9.6)	10.3–17.4 (14.3)	9.9–13.2 (11.2)	1.7–18.5 (12.8)	<loq-2.9 (2.3)</loq-2.9 	<loq-2 (1.5)</loq-2 	8.6	50
Copper	1.7–5.4 (3.6)	1.6–12.7 (6.3)	1.3–6.4 (3.4)	1.4–13.7 (5.8)	1-2.8 (2.1)	<loq-2.5 (2)</loq-2.5 	3.9	2,000
Iron	128.2–1563.5 (589.7)	59.9–3595.3 (891.2)	65.4–1610.5 (509)	19.6–554.7 (203.6)	12–536 (185.5)	19.9–414.7 (187.2)	423.1	200
Manganese	175.7–406.6 (285.2)	186.3–588.6 (319.2)	192.5–824.3 (388.4)	64.1–810 (292.6)	2.3–433.2 (133.6)	1.9–382.9 (133.9)	258	50
Nickel	5.2–8.4 (6.5)	6–8.3 (7)	3.4–5 (4.2)	4–6.5 (5.7)	2.2–3 (2.6)	3.8–4.3 (4)	5	20
Lead	<loq-1.7 (1.2)</loq-1.7 	0.81–7.1 (3.4)	<loq-1.1 (0.68)</loq-1.1 	<loq-1 (0.32)</loq-1 	<loq< td=""><td><loq-1.5 (0.58)</loq-1.5 </td><td>1.1</td><td>10</td></loq<>	<loq-1.5 (0.58)</loq-1.5 	1.1	10

Note that *Cm* is the average concentration considering all of the sampling campaigns. *RD 140/2003* represents the recommended limits for Spanish drinking water guidelines. *LOQ* limit of quantification

concentrations, especially in campaigns C1–C4, reaching maximum concentrations up to 0.032 mg/L (C3). The remaining metals (aluminium, copper, cadmium, chromium, etc.) were detected at low concentrations in all campaigns (C1–C6). Among these metals, average concentrations of iron, manganese and arsenic (0.012 mg/L) were above those established by the Spanish limit for drinking water (Table 2). The standard limits were exceeded in approximately 40% of the

sampled points for arsenic and iron and 90% of sampled points for manganese.

Finally, the average concentrations of the detected organic compounds were, on average, much lower than those from metals (Table 3). Among the 26 pesticides, 22 were not detected at any sample, two were detected only at one sample (over 23) and 4 were frequently detected. These four pesticides were atrazine, diazinon, TBA and terbutryn and all were

Table 3Concentration ranges and average concentrations (*in parentheses*) of pesticides and chlorinated solvents for each sampling campaign (C1–C4)in $\mu g/L$

Chemical parameter	Sampling campai	Cm	RD 140/2003			
	C1	C2	C3	C4		
Atrazine	0	0-0.037	0–≤LOQ (0.002)	0-0.011	0.008	0.1
Diazinon	0	0-0.012 (0.006)	0-0.03 (0.008)	$0 \le LOQ$ (0.001)	0.004	0.1
TBA	0.012–0.061 (0.041)	0.047–0.096	<loq-0.042 (0.016)</loq-0.042 	0-0.026	0.034	0.1
Terbutryn	0	0.056-0.12 (0.088)	0-0.022 (0.0053)	0–≤LOQ (0.007)	0.024	0.1
Total pesticides	0.012–0.061 (0.041)	0.17–0.24 (0.19)	<loq-0.01 (0.033)</loq-0.01 	<loq-0.041 (0.023)</loq-0.041 	0.072	0.5
PCE	0.58–1.98 (1.22)	1.06–5.13 (2.43)	1.51–5.25 (2.77)	0.42–44.2 (10.1)	4.13	-
TCE	0–1.25 (0.48)	1.17–4.14 (2.49)	0.59-4.74 (1.96)	0.34–2.1 (1.1)	1.5	-
PCE+TCE	0.58–3.23 (1.7)	2.23–9.2 7 (4.92)	2.14–9.99 (4.73)	0.78–44.8 (11.2)	5.63	10

RD 140/2003 is the recommended limits for Spanish drinking water guidelines. LOQ limit of quantification

detected at very low concentrations $(10^{-5}-10^{-6} \text{ mg/L})$. Somewhat high concentrations were found for the chlorinated solvents PCE and TCE, reaching values up to 0.043 and 0.004 mg/L, respectively. The average concentrations of the pesticides and the sum of PCE and TCE were below the limit established by the Spanish limit for drinking water (Table 3). The sum of the average concentration of both chlorinated solvents only exceeded the limit of 0.01 mg/L in C4.

To summarize, the chemical parameters that pose some risk to the use of groundwater as a drinking water source in descending order were ammonium, manganese, iron and arsenic. Moreover, chloride and sulphate also exceeded the limit set by Spanish drinking water guidelines in the sampling campaigns C1–C3. Among these compounds, ammonium significantly exceeded the limit set by the Spanish drinking water guidelines by one order of magnitude.

River-groundwater interaction

The chemical composition of the river varies over the time depending on the sources that discharge into the river. In the final part of the catchment, river water is a mixture of different waters such as raw sewage water, treated sewage water, industrial discharges, rainfall, etc. The industrial pollution is reflected in the presence of metals (e.g., manganese, iron and aluminium) and chlorinated solvents (PCE and TCE) in the river. High concentrations of some metals (e.g., iron, manganese, nickel and zinc) were found in the River Besòs and of PCE+TCE in the aquifers located in the central part of the catchment that is highly industrialized (Navarro and Carbonell 2007). Inorganic wastewater indicators (i.e., boron, nitrate and ammonium) as well as a wide array of organic pollutants such as surfactants (Tubau et al. 2010) and personal care products (PCPs; Serra-Roig et al. 2016) are also detected

in the river because some of them are not efficiently removed in WWTPs (e.g., Molins-Delgado et al. 2015, 2016). Changes in river-water quality influence the groundwater quality and, therefore, understanding the interaction between groundwater and surface water is a key issue to assess the potential uses of groundwater, especially in heavily pumped aquifers, where rivers are the main source of aquifer recharge.

Figure 5 shows the average concentrations in the River Besòs versus the average concentrations in the aquifer for major ions, metals and some organic compounds (e.g., pesticides and chlorinated solvents) from 2007 to 2014. Moreover, some PCPs detected in previous studies are also displayed (Serra-Roig et al. 2016). It can be observed that most of the major ions presented similar concentrations in the river than in the aquifer, except for nitrate (14.9 vs. 4 mg/L). Redox indicators (TOC, dissolved oxygen and ammonium) presented higher concentrations in the river than in the aquifer. The high average concentration of ammonium (5.1 mg/L), the low average concentration of dissolved oxygen (0.4 mg/L) and the presence of some dissolved metals provided evidenced of the reducing character of the aquifer. In fact, average concentrations of arsenic, iron and manganese were higher in the groundwater than in the river (Fig. 5). Iron and/or manganese hydroxides have a strong affinity for arsenic in aquifers (Mohan and Pittman 2007). Arsenic can be absorbed in these hydroxides and be released to some extent to groundwater, together with iron, when the reduction of the hydroxides occurs. This last observation could explain the increase of iron, manganese and arsenic when river water infiltrates the aquifer. Figure 6 shows that reduced arsenic, As (III), was the predominant dissolved species at some points (ADS-6n and SAP-2b) and had an important contribution in others (SAP1, SAP-2 and ADPW). These reducing conditions might support the natural attenuation of some of the organic compounds in the

Fig. 5 Average concentrations in the River Besòs and in the aquifer for several species such as major ions, redox indicators, pesticides, chlorinated solvents and some personal care products (*PCPs*, Serra-Roig et al. 2016)



Average concentration of the River Besòs

Fig. 6 On the *left axis*, As concentrations in groundwater samples collected in *C4– C6*. The ratio between As (III) and total As is plotted on *the right axis*



aquifer because their average concentrations in groundwater were lower than those of the river (Fig. 5). Conversely, there were other compounds such as the pesticides atrazine, terbutryn and TBA, both chlorinated solvents (PCE and TCE) and two PCPs (4MBC and BZT), that presented higher average concentrations in the aquifer than in the river; however, they were detected at low concentrations and, therefore, they are not a major concern to define the potential uses of groundwater.

Temporal evolution of groundwater quality

Groundwater composition varies according to the seasonal changes in river-water quality; therefore, the resident water composition in the aquifer is a mixture of the aforementioned river end-members (W1, D1 and D2) in different proportions (mixing ratios). The composition of these end-members and the measured versus computed concentrations are summarized in Table S1 and Figure S3 of the ESM.

Among these three river end-members, the dry season endmember D2 (57.3%) is the largest contributor to the groundwater recharge, followed by the river end-members W1 (32.2%) and D1 (10.5%). It is important to notice that dry season end-members dominated over wet season endmember but in a lower proportion than previous studies. Tubau et al. (2014) reported a proportion 4:1, whereas in the present study the proportion is 3:1 (dry end-members over wet end-member), which can be explained by the low contribution of the W1 end-member in the first three sampling campaigns (18.5, 20.3 and 21.9% for C1, C2 and C3, respectively) compared with the dry end-members (Jurado et al. 2015). In fact, these sampling campaigns were carried out when a severe drought period occurred from July 2007 to May 2008 and groundwater levels sharply decreased (Fig. 2). After this period, the groundwater levels progressively began to recover, and their oscillations depend mainly on the River Besòs flow dynamics and also on the continuous pumping in the parking area; therefore, the contribution of the W1 end-member was more significant in the last three sampling campaigns, being 41.1, 44.3 and 44.9% in C4, C5 and C6, respectively. These results were consistent with the rainfall events that occurred in the Besòs River Delta in May 2010 and December 2013.

To sum up, groundwater quality is highly dependent on the precipitation events that occurred in the Besòs River Delta catchment because it was less mineralised when the contribution of the wet end-member was higher than 40% of the total recharge (from C4 to C6). Therefore, groundwater presented better quality when a significant part of the groundwater recharge corresponds to rain water. In fact, as pointed out before, most of the species considered had lower average concentrations in the last three sampling campaigns (C4–C6) than those in the first sampling campaigns (C1–C3) (e.g., chloride, sulphate, sodium, bicarbonate, arsenic and iron; Tables 1 and 2).

Potential uses of the pumped urban groundwater

It is clear that there is a large volume of pumped urban groundwater available in the study area. Unfortunately, groundwater quality does not meet the Spanish drinking water standards for being used as potable water because there are some parameters that exceeded the limit set by the Spanish drinking water guidelines (RD 140/2003). The major threats that were found in the urban groundwater are ammonium and the metals manganese, iron and arsenic. Taking into account that the daily water usage per person is 200 L (73,000 L/year), the volume of 3×10^{6} m³ will allow supplying water to the whole city of Sant Adria del Besòs in stressful situations such as drought periods. However, the pumped groundwater would need additional treatment to make it suitable for drinking purposes, which can be achieved by remediation measures that have to be applied to the pumped groundwater-for example, groundwater quality could be improved by recharging the pumped groundwater via an artificial aquifer recharge scheme using ponds or wells (which can be constructed upstream). The artificial recharge would allow improving the regional groundwater quality; moreover, this method is efficiently used for drinking water production because several studies have pointed out that artificial recharge reduces the concentration of metals, ammonium, organic matter and organic contaminants (Bekele et al. 2011; Haeffner et al. 2001; Valhondo et al. 2015). Another remediation approach is pump-and-treat, which involves the treatment of the water and re-injecting it into the aquifer. Such technology has been employed to remove iron from groundwater but it is expensive (US EPA 2001). Other possible groundwater treatment techniques include physicochemical treatment options (i.e., aeration, ion exchange, breakpoint chlorination and reverse osmosis) and biological filters. Aeration can promote the removal of dissolved metals and ammonium from groundwater through oxidation (Siabi 2008). Further treatment (sedimentation and filtration) is required after aeration when the amount of iron is high and when manganese is present (Vigneswaran et al. 2007). Ammonium can also be efficiently removed from groundwater by ion exchange processes using zeolites, by adding chlorine to water and by biological filters where ammonium is oxidized (to nitrite and then to nitrate) by different bacteria (Groeschke et al. 2016). Overall, biological removal is preferable because there is no need for the addition of extra chemicals (e.g., chlorine) to the treated water (Tekerlekopoulou and Vayenas 2007).

The pumped groundwater is of sufficient quality to be used for cleaning streets and for watering the public parks and gardens. In fact, currently, less than 1% of this groundwater is used for watering gardens and cleaning the streets. Other realistic potential uses of this resource in the Besòs River Delta can be: (1) supplementing the river flow for ecological benefit, especially in the summer season and/or (2) mitigating seawater intrusion problems through hydraulic barriers. Currently, the aquifers of the Besòs River Delta do not have seawater intrusion problems (Vázquez-Suñé et al. 2016) but a barrier has recently been constructed using treated wastewater to prevent seawater intrusion in the main aquifer of the neighbourhood Llobregat River Delta (Ortuño et al. 2012).

The water management authorities, together with the city council, must evaluate the cost-benefits to decide whether this valuable resource can be used for supplying the inhabitants of Sant Adrià del Besòs (e.g., drinking purposes) and/or environmental purposes (e.g., to support ecologically the river flow or urban irrigation).

Conclusions

steps: (1) to quantify the groundwater recharge, (2) to characterize the sources of groundwater contamination, (3) to assess the temporal evolution of groundwater quality, and (4) to propose the potential uses of the pumped urban groundwater. This approach has been applied to the shallow aquifer of the Besòs River Delta in the city of Sant Adrià del Besòs. The results show that the volume of river water that infiltrates the aquifer is sufficient to supply the whole city of Sant Adrià del Besòs. The main recharge source of the aquifer is a polluted river, which controls the groundwater quality. Groundwater quality is less mineralized when a significant part of the groundwater recharge comes from rain water (i.e., wet river end-member contribution up to 40% of the total recharge). The limiting factor to propose the potential uses of this resource is its quality because there are some parameters such as ammonium, manganese, iron and arsenic, that exceeded the limit set up by the Spanish drinking water guidelines (RD 140/2003). As a result, this urban groundwater cannot be used as potable water before being further treated. Other potential uses could be supplementing the ecologically important river flow and/or mitigating seawater intrusion problems through hydraulic barriers. Currently, it is only used for watering plants and cleaning public areas of the city. This work shows that groundwater may reduce pressure on conventional freshwater supply resources in urban areas in moments of high demand (e.g., drought periods) but there is an urgent need for proper management of this resource. Usually, little consideration is given to pumped groundwater resources in urban areas because it has not been used in the past, or present water use is limited, or even because the groundwater potential is neglected. The presented approach can be applied to other aquifers with similar purposes.

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